Acid Rain Research

REPORT 7/1985

RAIN project. Annual report for 1984.

Norwegian Institute for Water Research NIVA
Environmental Engineering Division

NIVA - REPORT

Norwegian Institute for Water Research NIVA

Royal Norwegian Council for Scientific and Industrial Research

Address: Postbox 333, Blindern, Oslo 3, Norway

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Report No.: 0-82073

Sub-No.:

Serial No.: 1707

Limited distribution:

Report title: RAIN project. Annual report for 1984

Date: February 1985

Project No: 0-82073

Topic group: Acid precipitation

Geographical area: Norway

Number of pages (incl. app.)

Author(s): Richard F. Wright

Contractor:

Contractors ref. (or NTNF - No):

Abstract: Project RAIN is a 5-year international research project aimed at investigating the effect on water chemistry of changing acid deposition to whole catchments. The project comprises 2 parallel large-scale experimental manipulations - artificial acidification at Sogndal and exclusion of acid rain at Risdalsheia. The project began in June 1983. The first year was devoted to selection of sites, collection of background data on precipitation, runoff and soils, and design and construction of roofs, watering systems, weirs and sampling devices.

Treatment at Sogndal commenced in April 1984 with the acidification of the snowpack by addition of sulfiric acid (catchment 2) or a 1:1 mixture of sulfuric and nitric acids (catchment 4). Treatment continued in August-October 1984. Preliminary results indicate rapid and significant response in runoff chemistry to the acid treatment; pH declines (to as low as 4.1 during snowmelt), SO_4, NO_3 and labile Al increase.

At Risdalsheia treatment began in June 1984 with the mounting of transparent panels on the roofs at KIM catchment (treatment by deacidified rain) and EGIL catchment (control with ambient acid rain). Preliminary data up to 15 October 1984 indicate a decrease in nitrate concentration in runoff from KIM catchment.

4 keywords, Norwegian	4 keywords, English
1. Sur nedbør	1. Acid precipitation
2. Vannkjemi	2. Water chemistry
3. Reversibilitet	3. Reversibility
4. Nedbørfelt	4. Catchments

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ISBN 82-577-0891-7

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RAIN project

Annual report for 1984

Oslo, February 1985

Prepared by Richard F. Wright
Project RAIN Manager

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PREFACE

A large number of individuals and institutes have cooperated in the RAIN project during 1984. The project scientists include N. Christophersen, E. Gjessing, E. Lotse, H.M. Seip, A. Semb and R.F. Wright. Technical staff includes S. Andersen, R. Høgberget, A. Rogne, B. Slettaune, R. Storhaug and K. Wedum. NILU, NIVA, SI, and the Department of Soil Sciences, SLU provided technical support.

The RAIN project would not be possible without the generous cooperation of landowners at both sites. We thank N. Knagenhjelm, Sogn Televerk and Arendal Televerk for permission to use private roads. N. Dalaker, H. Haukås, A. Meltveit and A. Risdal provided local assistance.

Financial support in 1984 came from the Norwegian Ministry of Environment, The Royal Norwegian Council for Scientific and Industrial Research, the Ontario Ministry of the Environment, Environment Canada, The Swedish National Environmental Protection Board, and the Surface Water Acidification Project (SWAP) (The Royal Society, the Norwegian Academy of Science and Letters, and the Royal Swedish Academy of Sciences).

1. INTRODUCTION

Project RAIN (Reversing Acidification in Norway) is a 5-year international research project. The project comprises two parallel large-scale experimental manipulations with natural headwater catchments to determine the response of runoff chemistry to changes in the loading of strong acids from the atmosphere (Figure 1). The project will provide information on reversibility of acidification, rate of response and target loadings.

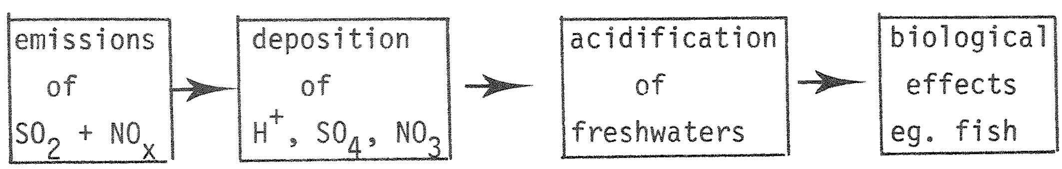


Figure 1. The chain of events linking emissions to the atmosphere to adverse biological effects. Project RAIN focuses on the link between deposition and freshwater acidification.

Experiment 1: Acidify two pristine catchments by adding a small volume of highly acidic rain to natural precipitation. At Sogndal, western Norway, (Figure 2) catchment 2 is being acidified by sulfuric acid while catchment 4 receives a 1:1 mixture of sulfuric and nitric acids.

Experiment 2: "Restore" an acidified catchment by excluding ambient acid precipitation and watering with clean precipitation beneath the roof. At Risdalsheia, southernmost Norway, (Figure 2) KIM catchment receives clean rain beneath the roof, while EGIL catchment receives recycled ambient acid rain.

At both sites one or more additional catchments serve as controls.

The 5-year project plan calls for approximately 1 year of pre-treatment data, 3 years of treatment, and 1 year of post-treatment recovery.

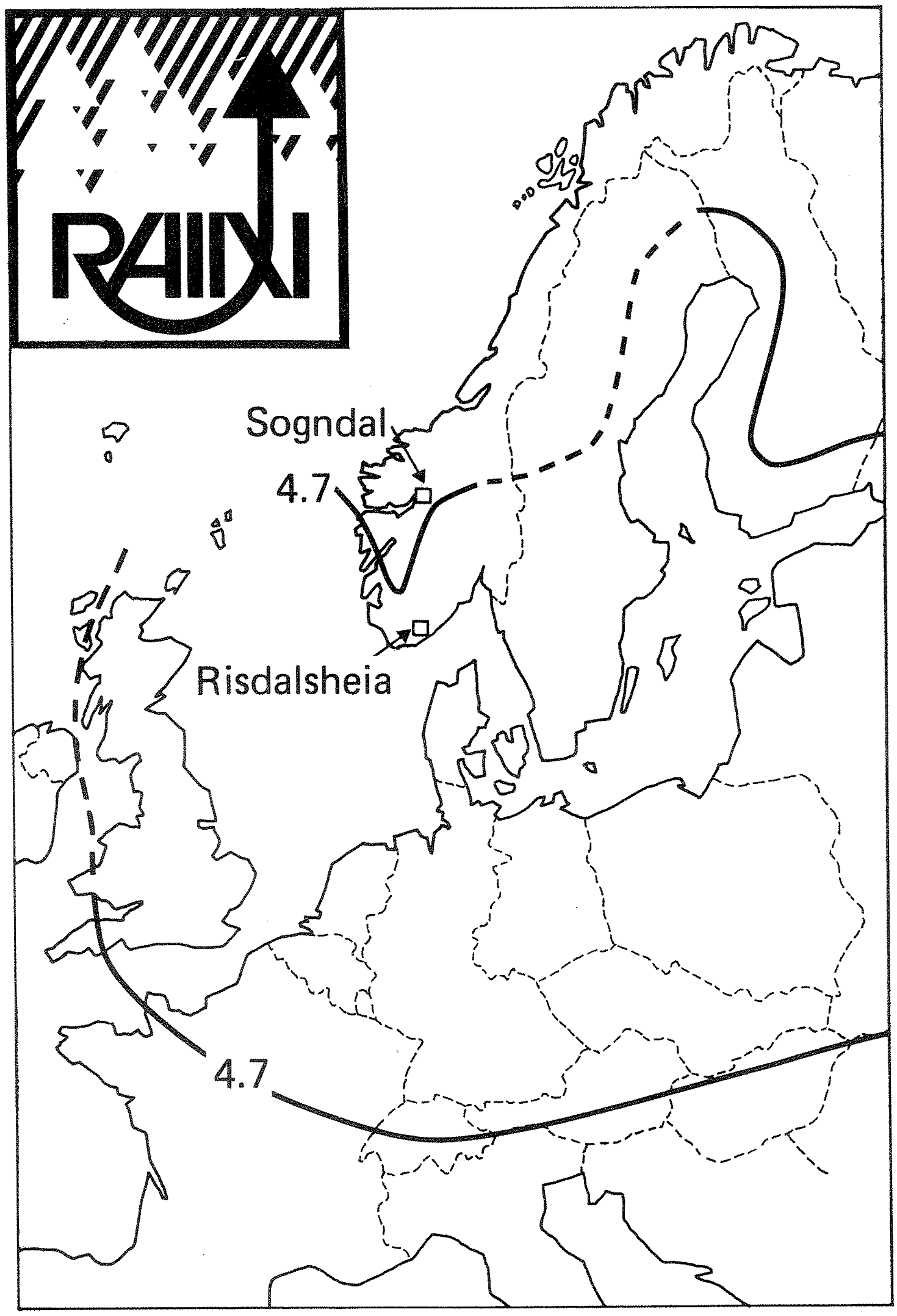


Figure 2. Location of the experimental catchments in project RAIN. Areas within the pH 4.7 isoline receive precipitation with a yearly weighted average pH below 4.7.

Project RAIN is conducted jointly by scientists from:

- Norwegian Institute for Water Research (NIVA)
- Norwegian Institute for Air Research (NILU)
- Center for Industrial Research (SI)
- Norwegian Water Resources and Electricity Board (NVE)
- Department of Soil Science, Swedish Universty for Agricultural Sciences (SLU).

The project receives financial support from:

- Norwegian Ministry of Environment
- Royal Norwegian Council for Scientific and Industrial Research
- Ontario Ministry of the Environment
- Environment Canada
- Swedish National Environmental Protection Board
- Surface Water Acidification Project (SWAP) (The Royal Society, The Norwegian Academy of Science and Letters and the Royal Swedish Academy of Sciences).

Additional support comes from in-house funds of the participating institutes.

Project RAIN began in June 1983 following a pilot study conducted in 1980 and subsequent planning. The activities in the first 10 months of the project (1 June 1983 - 31 March 1984) are summarized in the first annual report. Major accomplishments were:

- selection of experimental sites and signing of contracts with landowners

- design and construction of roofs, watering systems, weirs and sampling devices

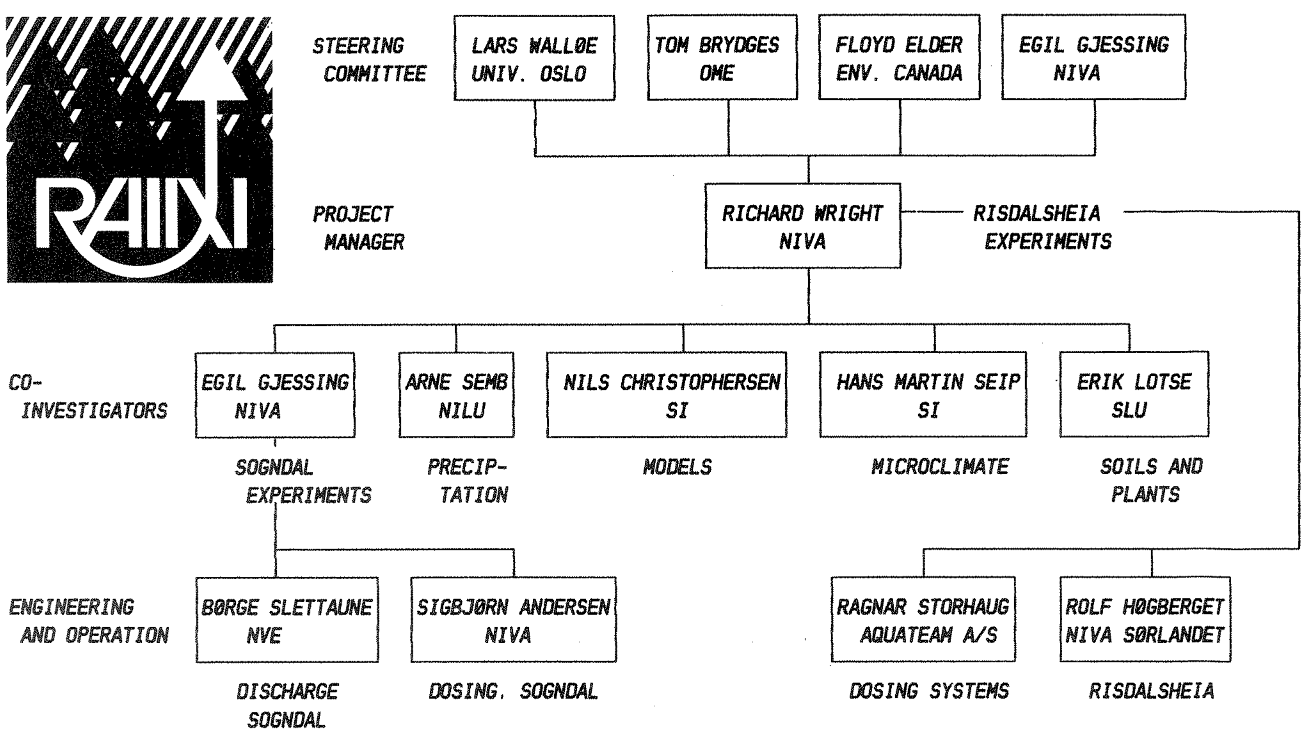
- collection of pretreatment data for precipitation, runoff and soils.

This report describes project activities and accomplishments during 1984.

2. PROJECT ORGANIZATION

The organization structure of Project RAIN cosist of a steering committee, a project manager, a scientific staff and a technical staff (Figure 3). The project is coordinated by NIVA.

PROJECT RAIN ORGANIZATION CHART 1 JANUARY 1985



NILU Norwegian Institute for Air Research
NIVA Norwegain Institute for Water Research
NVE Norwegian Electricity and Water Resources Board
OME Ontario Ministry of the Environment
SI Central Institute for Industrial Research
SLU Swedish University for Agricultural Sciences

Figure 3. Organization chart for Project RAIN (as of January 1985).

3. DESCRIPTION OF SITES

3.1. Sogndal

For the acidification experiments four catchments have been selected at Sogndal (Figure 4). The site is located above treeline at about 900 m above sea level. The vegetation is alpine and includes dwarf birch, heather, grasses, mosses and lichens. Soils are thin and poorly-developed, (US classification: lithic haplumbrept, sandy, siliceous, frigid). Soil pH (H_2O) is 4.5-5.5. Bedrock is siliceous gneiss.

Sogndal

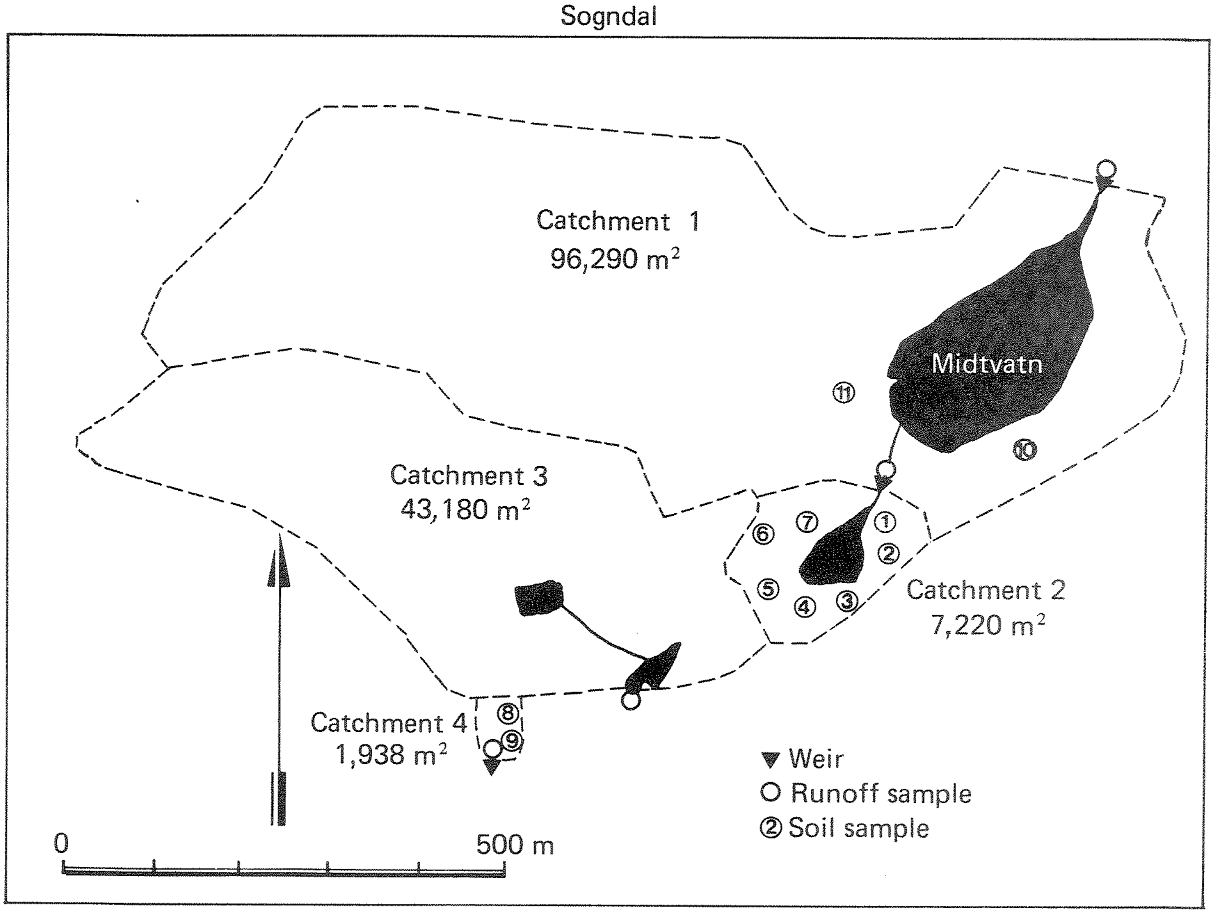


Figure 4. Overview map of the 4 catchments at Sogndal. Catchments 1 and 3 are controls while catchment 2 receives H_2SO_4 and catchment 4 H_2SO_4+HNO_3.

Precipitation at Sogndal is relatively clean with a volume-weighted average pH of 4.9 and excess sulfate concentration of 14 µeq/l (Table 1). Runoff chemistry from the four catchments prior to treatment indicates extremely sensitive systems with pH 5.5-6.0, Ca concentrations 10-30 µeq/l and SO_4 15-25 µeq/l (Table 1).

Table 1. Volume-weighted mean concentrations of major ions, aluminium species and organic carbon in precipitation (weekly bulk samples at Haukås farm) and runoff (Catchment 3 control) at Sogndal for the period 831017-841021. Runoff data are preliminary pending calibration of weirs. Units: µeq/l unless otherwise indicated.

Parameter	Precipitation n=45 total	excess	Runoff n=36 total	excess
H_2O (mm)	1045		-	
H^+	14	14	2	2
(pH)	(4.86)		(5.8)	
Na	64	2	60	9
K	6[a]	6[a]	2	2
Ca	7	4	21	19
Mg	14	0	14	2
NH_4	7	7	1	1
NO_3	6	6	1	1
Cl	72	0	60	0
SO_4	21	14	24	18
HCO_3	0	0	7	7
Sum +	112	33	100	33
Sum -	99	20	92	26
Tot-Al µg/l	-	-	29	-
R-Al µg/l	-	-	16	-
NLAL µg/l	-	-	10	-
TOC mg/l	-	-	1	-

[a] for the period 830620-840610.

3.2. Risdalsheia

For the de-acidification experiments three natural headwater catchments have been selected at Risdalsheia, located across the Tovdal River from the long-term calibrated catchment at Birkenes (Figures 2 and 5). The area is 300 m above sea level. The vegetation at Risdalsheia is similar to that at Sogndal except that here pine and spruce are also present. The soils are also similar - thin, patchy and poorly - developed. A major difference is that here soil pH (H_2O) is about 1 unit lower (pH 3.9-4.5). Bedrock is biotite granite.

Risdalsheia lies in the zone of maximum deposition of acid components in Norway. Data from the nearby Birkenes station show a long-term volume-weighted average pH of 4.2 in wet precipitation (SFT 1984). Deposition of SO_4 is 70 kg/ha·yr (50 wet plus 20 dry).

Runoff from the three catchments prior to treatment was highly acidic (pH 3.9-4.2) with high levels of sulfate and inorganic labile aluminium (Table 2).

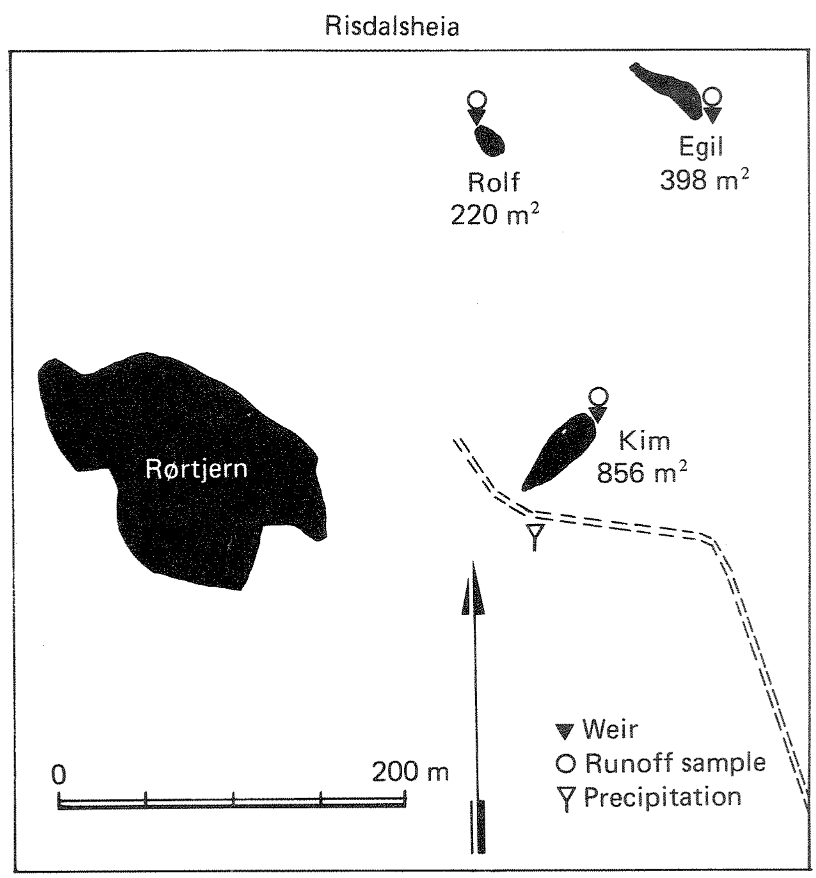


Figure 5. Overview of the 3 catchments at Risdalsheia. KIM and EGIL are covered by roofs; ROLF is reference. Water supply for snowmaking is the pond Rørtjern.

Table 2. Volume-weighted mean concentrations of major ions, aluminium species and organic carbon in precipitation (daily samples at Birkenes) and runoff (EGIL-catchment) at Risdalsheia. Precipitation data are for the calendar year 1983 (SFT 1984). Runoff data are for the period 840802-841018. Units; μeq/l unless otherwise indicated.

Parameter	Precipitation n=134 total	Precipitation n=134 excess	Runoff EGIL n=17 total	Runoff EGIL n=17 excess
H_2O (mm)	1313		-	
H^+	47	47	99	99
(pH)	(4.33)		(4.00)	
Na	51[a]	0	64	11
K	-	-	3	3
Ca	12	10	11	9
Mg	14	2	16	4
NH_4	36	36	3	3
NO_3	35	35	19	19
Cl	59	0	62	0
SO_4	65	57	119	113
HCO_3	0	0	0	0
Sum +	160	95	196	129
Sum -	159	92	200	132
Tot-Al μg/l	-	-	440	-
R-Al μg/l	-	-	370	-
NLAL μg/l	-	-	230	-
TOC mg/l	-	-	10.9	-
F μg/l	-	-	35	-

[a] estimated from Cl concentration.

4. TECHNICAL ASPECTS

4.1. Sogndal

At Sogndal 3 of the 4 catchments are equipped with weir and level recorder for continuous gauging of discharge (Table 3). Installation, maintenance and acquisition of discharge data are carried out by the Norwegian Water Resources and Electricity Board (NVE). Discharge measurement began in August 1983 (December 1983 at catchment 1).

Table 3. The Sogndal catchment. Treatment, area and measurement program.

	Catchment			
	1	2	3	4
Treatment	Control	H_2SO_4	Control	H_2SO_4+HNO_3
Catchment area	96290 m^2	7220 m^2	43180 m^2	1938 m^2
Discharge	from 841215	from 830816	no weir	from 830816
Runoff chemistry	from 830608	from 830608	from 830608	from 830608
Precipitation chemistry	At Haukås farm from 830607			

A precipitation collector for weekly bulk samples is located at the Haukås farm (500 m above sealevel) about 3 km east of the experimental site. Precipitation chemistry sampling began in June 1983. Parameters measured include volume, pH, conductivity, Na, K, Ca, Mg, NH_4-N, NO_3-N, SO_4 and Cl. Precipitation sampling and analysis is conducted by the Norwegian Institute for Air Research (NILU).

Samples of runoff for chemical analysis are collected weekly at each of the 4 catchments. Sampling began in June 1983. Parameters measured include pH, conductivity, Na, K, Ca, Mg, NH_4-N, NO_3-N, SO_4, Cl, F, alkalinity, total-Al, acid-reactive Al, non-labile Al and total organic carbon. Samples are collected more frequently during treatment. Samples are sent by mail to the Norwegian Institute for Water Research (NIVA) for analysis.

Soils at Sogndal were sampled in August 1983 and again in August 1984 (Figure 4). Vegetation samples were collected in August 1984. Soil and vegetation studies are conducted by Prof. Erik Lotse, Department of Soil Sciences, Swedish University for Agricultural Sciences, Uppsala. In addition to description of the soil profiles analyses include pH, loss on ignition, C, N, S, exchangeable acidity, exchangeable cations (Na, K, Ca, Mg, NH_4, H, and Al), base saturation, and cation exchange capacity.

Treatment at Sogndal commenced in April 1984 with the application of acid to the snowpack of catchments 2 and 4. Catchment 2 received a total of 5 l concentrated H_2SO_4 (corresponds to 0.37 g S/m^2) (Table 4). Catchment 4 received 0.7 l conc. H_2SO_4 plus 1.525 l conc. HNO_3 (corresponds to 0.19 g S/m^2 and 0.52 g N/m^2) (Table 4). The acid was diluted to about 0.5 M (corresponds to about 180 l of solution for catchment 2) and sprayed on the snow by means of a conventional backpack insecticide tank with hand-held spray pump. The acid was spread evenly over the entire catchment. The applied volume corresponded to about 0.025 mm precipitation.

Additional acid was applied four times during the late summer and autumn 1984 (Table 4). Catchment 2 received 8.64 l conc. H_2SO_4 in 4 equal portions while catchment 4 received 1.16 l conc. H_2SO_4 plus 1.96 l conc. HNO_3 in 4 equal portions. The acid was applied by means of a conventional sprinkler system used in agriculture marketed under the trademark Wright Rain. At catchment 2 a tractor-driven pump supplied about 280 l/minute of water from Lake Midtvatn. (The weir at catchment 1 is at the outlet of Lake Midtvatn). This corresponds to approximately 2 mm/hour. The acid is diluted to about 50 l stock solution which is then metered into the main water line. The mixture has a pH of about 3.2. A total of 12 sprinklers were used to cover the 7220 m^2 of catchment 2. The watering procedure calls for first the application of about 2 mm of untreated water, then application of 11 mm of pH 3.2 water, and finally about 2 mm of untreated water. This procedure insures that the vegetation is wetted prior to application of acid, and that the acid is washed off at the end of treatment.

Plate 1. Sogndal. Application of acid to the snowpack in April 1984. Poles in the distance mark the catchment boundary.

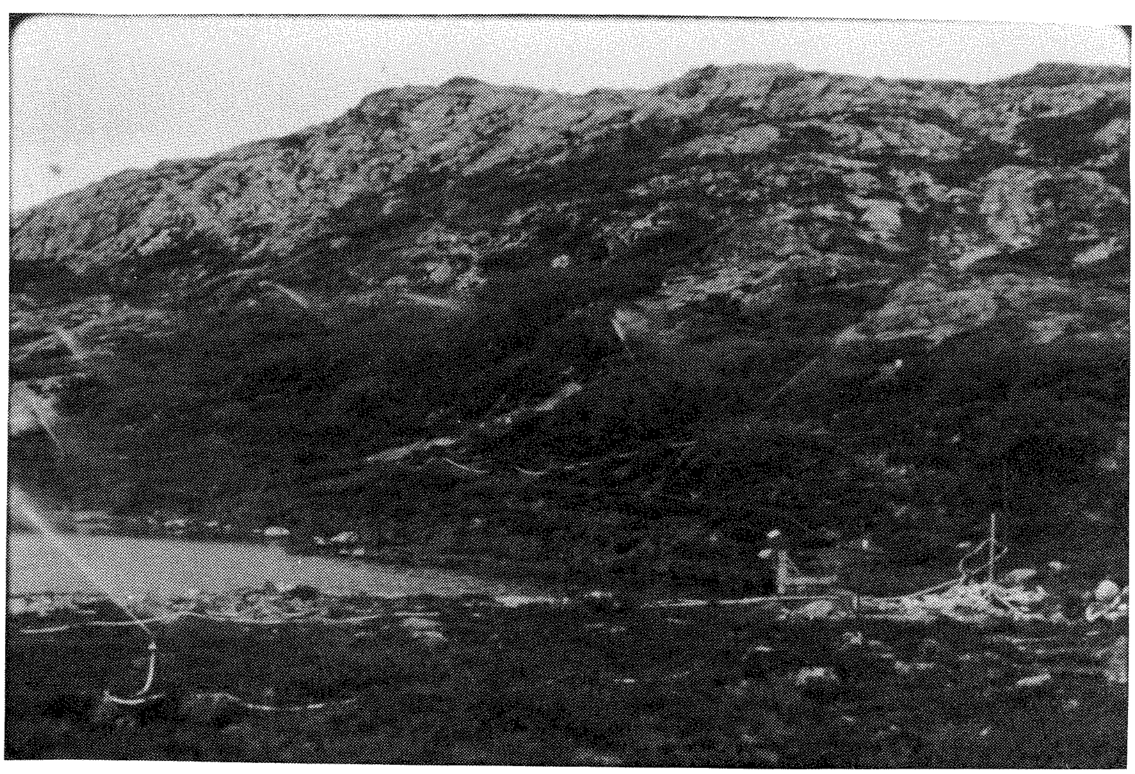


Plate 2. Sogndal. Application of acid to catchment 2 in September 1984. The weir at outlet to the small pond is at lower right.

Table 4. Acid doses applied to catchments 2 and 4 at Sogndal in 1983-84 (831017-841021). Shown for comparison are the loadings (wet + dry) at Birkenes and Storgama catchments in southernmost Norway (Christophersen et al. 1982, Christophersen et al. 1983). Units: keq/km^2.

Catchment	H_2O (mm)		H^+		SO_4		excess-SO_4		NH_4		NO_3	
	2	4	2	4	2	4	2	4	2	4	2	4
Natural precip.	1045	1045	14.6	14.6	21.9	21.9	14.6	14.6	7.3	7.3	6.3	6.3
Added to snow	.02	.02	25.0	25.0	25.0	12.5	25.0	12.5	0	0	0	12.5
Added 1	16	14	11.2	11.2	11.2	5.6	11.2	5.6	0	0	0	5.6
during 2	15	14	11.2	11.2	11.2	5.6	11.2	5.6	0	0	0	5.6
summer 3	15	14	11.2	11.2	11.2	5.6	11.2	5.6	0	0	0	5.6
4	15	15	11.2	11.2	11.2	5.6	11.2	5.6	0	0	0	5.6
SUM	1106	1102	84.4	84.4	91.7	56.8	84.4	84.4	7.3	7.3	6.3	41.2
Birkenes (1972-78)	1403		137		142		137		53		53	
Storgama (1973-78)	1108		66		72		70		30		34	

4.2. Risdalsheia

At Risdalsheia the experiment includes 3 natural headwater catchments (Figure 5, Table 5). Two catchments (KIM-treatment; EGIL-control) are covered by roofs and one catchment (ROLF-reference) serves as untreated reference. The roofs extend 2-3 meters beyond the catchment boundaries.

Table 5. The Risdalsheia catchments. Area, treatment and measurement program.

Treatment	KIM roof, "clean" rain	EGIL roof, acid rain	ROLF no roof, acid rain
Catchment area	856 m^2	398 m^2	220 m^2
Roof area	1165 m^2	650 m^2	-
Volume runoff tank	0.39 mm	0.93 mm	-
Discharge measured	from 840320	from 840320	from 841031
Runoff chemistry	from 831011	from 830905	from 830905
Precipitation chemistry	At Birkenes from 810701, at Risdalsheia from 841031.		

Runoff volume is gauged by an automatic system in which all water leaving each catchment is collected in a 500-l tank equipped with an automatic flushing valve and data logger. The system is designed and operated by NIVA. Discharge measurement began 20 March 1984 (at ROLF 31 October 1984). Samples for chemical analysis are collected from each tank by means of automatic sampler. Samples are sent by mail to NIVA for chemical analysis (pH, conductivity, Na, K, Ca, Mg, NH_4-N, NO_3-N, SO_4, Cl, F, alkalinity, total-Al, acid-reactive Al, non-labile Al, and total organic carbon). Sampling began in September 1983.

Weekly samples of bulk precipitation are collected at Risdalsheia and analyzed by NILU. These data supplement the nearby station at Birkenes, at which daily samples for air- and precipitation-chemistry are collected (also by NILU). In addition weekly samples are collected of precipitation applied beneath the roofs in KIM and EGIL catchments. Continuous measurement of temperature and humidity in KIM and EGIL catchments and outside the roofs are also conducted by NILU (since August 1984).

Soils at KIM and EGIL catchments were first sampled in October 1983. Soils and vegetation at KIM, EGIL and ROLF catchments were sampled in August 1984. Sample collection is designed such that integrated values for each catchment are obtained. Soil and vegetation studies are carried out by Prof. Erik Lotse, Department of Soil Sciences, Swedish University for Agricultural Sciences, Uppsala. In addition to description of soil profiles analyses include pH, loss on ignition, C, N, S, exchangeable acidity, exchangeable cations (Na, K, Ca, Mg, NH_4, H, Al), base saturation, and cation exchange capacity.

Roof construction at Risdalsheia began in January 1984. The construction contract was awarded to Opedal A/S, Langesund, after a round of bidding. The steel framework was set up during the winter using a crane and helicopter. The contract specified minimal disturbance of the catchments. The side panels are of corrugated fiberglass. The roof panels are clear acrylic and pass 85 % of the light in the photosynthetic range. The side panels are fitted with a cable and pulley system such that they are open during the summer half-year to permit access and adequate ventilation, and are closed during the winter to prohibit snow from blowing in under the rofs. The total cost of the two roofs was approximately NOK 1.8 million.

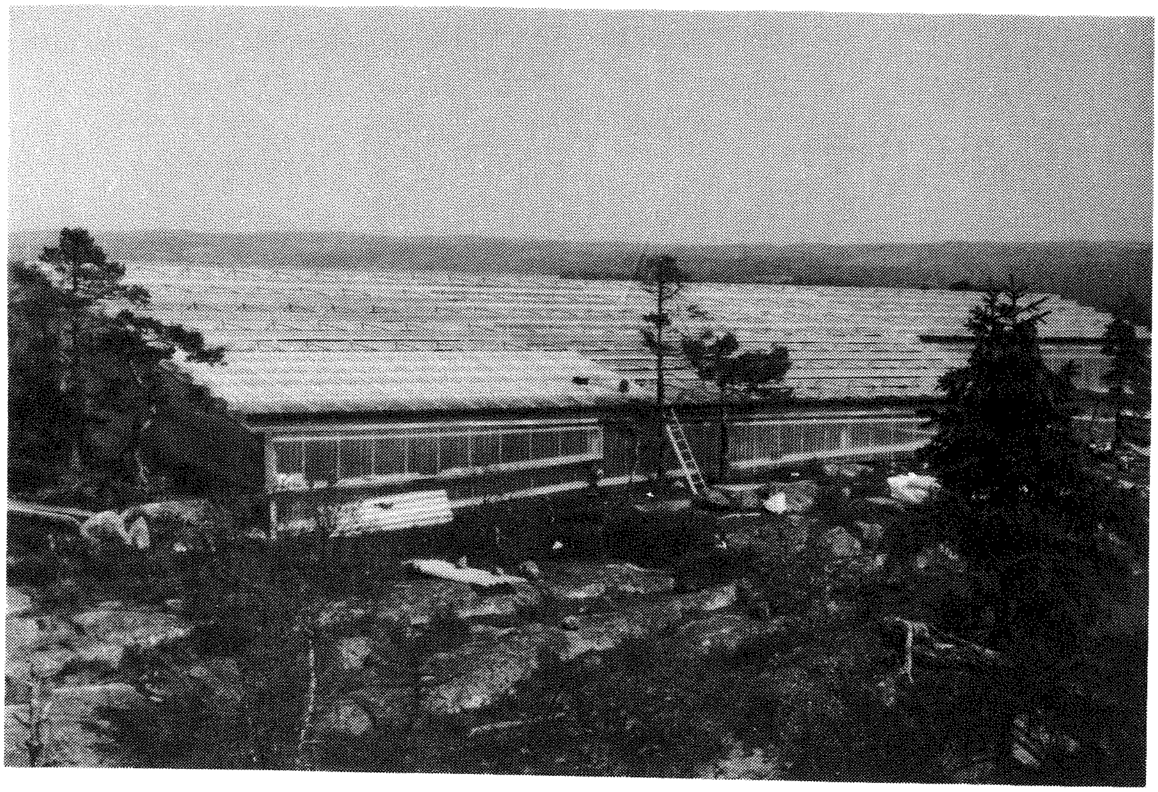


Plate 3. Risdalheia. Mounting of roof panels on KIM catchment in June 1984.

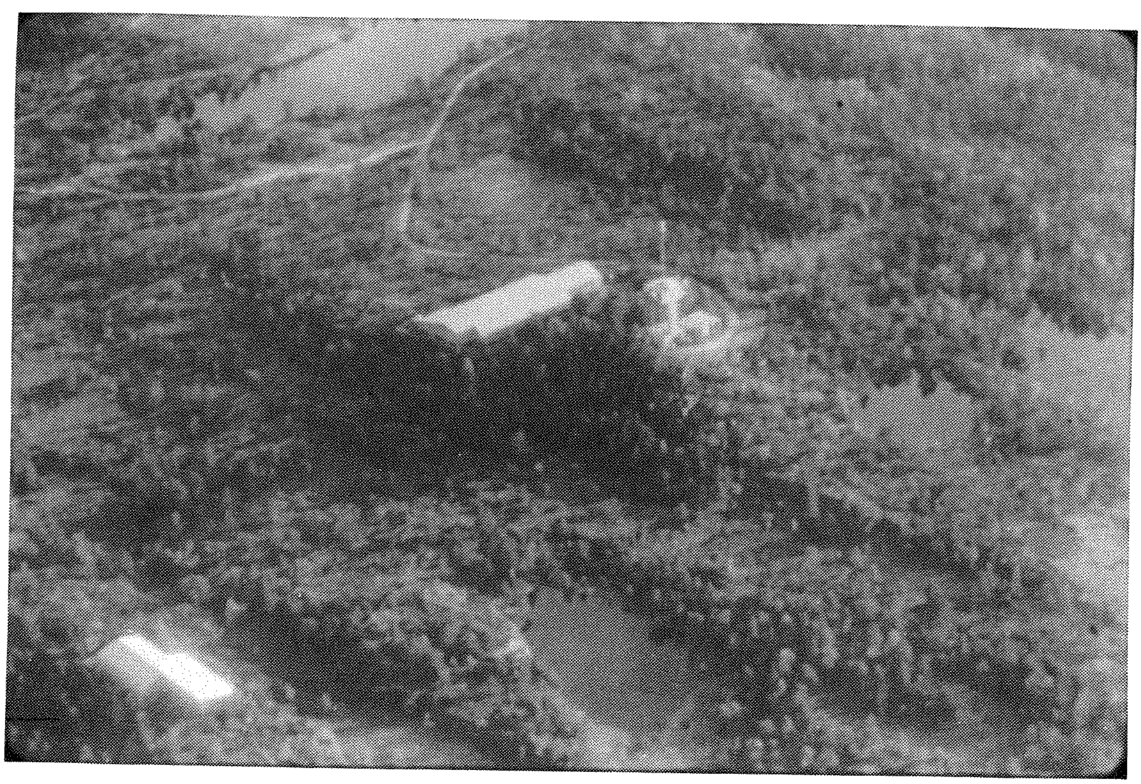


Plate 4. Risdalsheia. Aerial view of the 2 roofed catchments KIM (middle) and EGIL (lower left). Reference catchment ROLF is to the left of the small pond (bottom center).

The sprinkling systems collect incoming precipitation from the roofs by a gutter and cistern system. At KIM catchment the water is sent through one of two ion-exchange columns filled with mixed-bed resin. The ion-exchanged water has a conductivity of <2 µS/cm. Seawater is then added to the water in a ratio 1:8000 (summer) or 1:5000 (winter). The resulting precipitation has a chemical composition equivalent to that at Birkenes except that H^+, excess SO_4, NO_3 and NH_4 are removed. The water is pumped back out to a sprinkling system mounted under the roof. Watering proceeds at maximum 2 mm/hour. The entire system is automatic and regulated by the water level in the cisterns. The system at EGIL catchment is similar to that at KIM except that the water is not treated in any way, but merely recycled back under the roof.

Treatment at Risdàlsheia began 13 June 1984. As of 12 November 1984 490 m^3 precipitation had been treated at KIM catchment (corresponds to 405 mm) and 365 m^3 at EGIL catchment (corresponds to 550 mm). The difference reflects technical problems incurred during the first several months of treatment, including lightning damage and leaks in the gutter systems. The side panels were lowered and the sprinkling systems drained and closed down for the winter in early December 1984.

5. RESULTS

5.1. Sogndal

Precipitation at Sogndal had a volume-weighted average pH of 4.8 and excess SO_4 concentration of 15 µeq/l (Table 1). Episodes of acid precipitation occurred several times in 1983-1984. Bulk precipitation was as acid as pH 3.6 (29-31 August 1983, 6.1 mm). Winter precipitation contains 5 times higher concentrations of seasalts (100 µeq/l Cl) as compared with summer precipitation (15-20 µeq/l Cl).

The weirs at Sogndal have not yet been fully calibrated, and therefore discharge data are not yet available. The results from soil survey and analyses will be reported separately by E. Lotse.

Runoff chemistry at the control catchments 1 and 3 and the treated catchments 2 and 4 prior to treatment is characterized by pH 5.5-6.5, SO_4 concentrations 15-50 µeq/l, NO_3 concentrations 0-10 µeq/l, and labile-Al concentrations 0-10 µg/l. These are levels typical of sensitive waters in areas not receiving highly acidic precipitation (Wright 1983).

Treatment with acid resulted in immediate and dramatic changes in water chemistry. During snowmelt pH at catchment 2 (sulfuric acid) decreased to 4.09 accompanied by 180 µeq/l SO_4 and 300 µg/l labile-Al (Figure 6). The water chemistry recovered rapidly to background levels. Due to difficult access to the weir at catchment 4 no samples were collected during the critical snowmelt period 20 April - 14 May 1984; the effect of acidifying the snowpack with sulfuric plus nitric acids is thus unknown.

Both treatment catchments displayed water chemistry of composition similar to the control catchments during the summer 1984. The next treatment at the end of August resulted in response within hours at both catchments (Figure 6). Recovery upon cessation of treatment was also rapid.

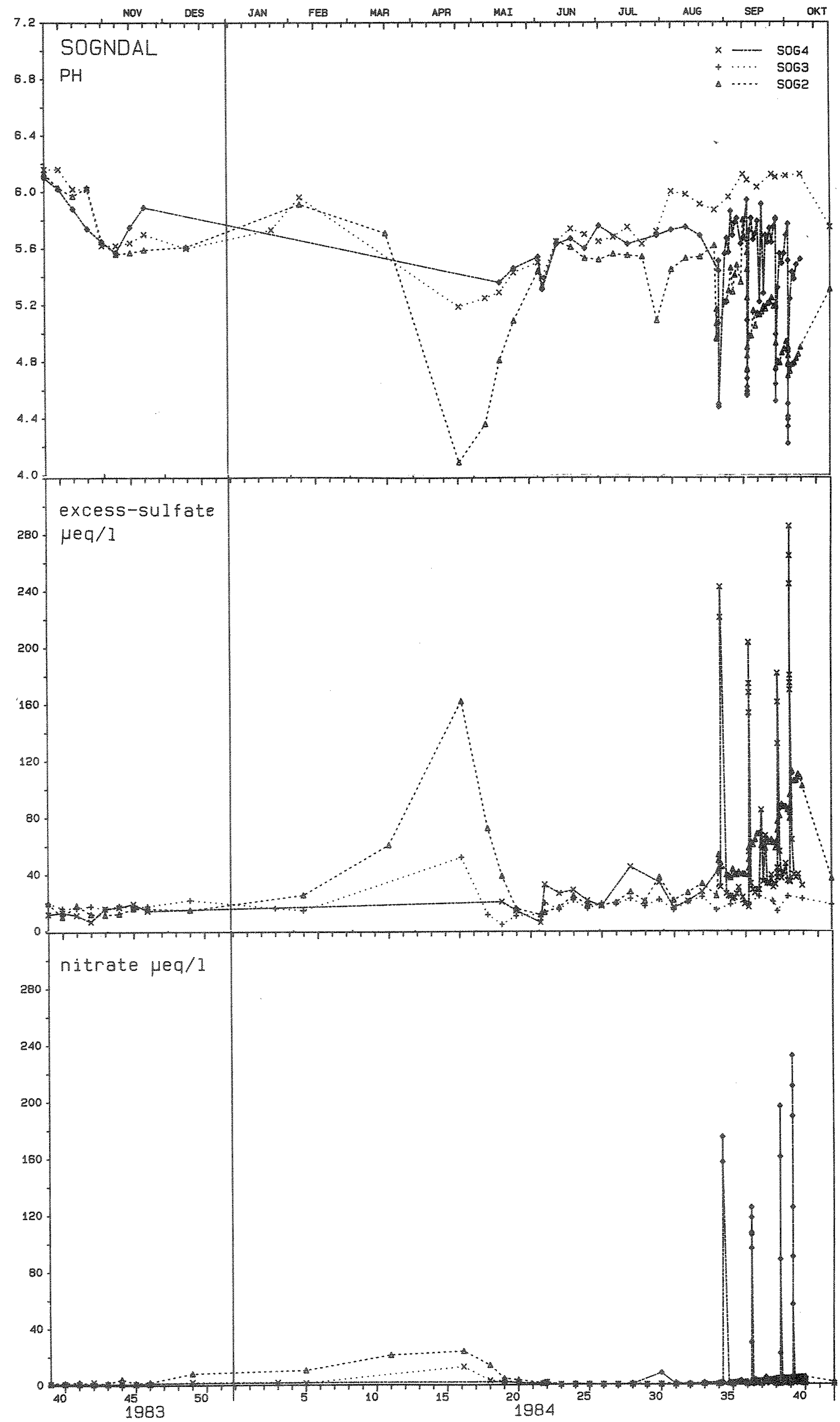


Figure 6. pH and concentrations of excess SO_4 and nitrate in runoff at catchment 3 (control), catchment 2 (H_2SO_4 treatment) and catchment 4 (H_2SO_4+HNO_3 treatment) at Sogndal for the first year of treatment.

Catchment 2 responds slower than catchment 4, problably because of the small pond at the bottom of catchment 2. This pond acts as a hydrologic buffer. For acidic water to leave catchment 2, the volume of pond water must first be flushed out.

The rapid response at both catchments to the acid treatment may be to a large degree due to "rain" landing on free water surfaces in the catchment. This pond and stream water may flow directly out of the catchment with only minimal contact with soil. A few percent of the applied pH 3.2 "rain" might thus be sufficient to bring about a drop in pH at the weir 4.5. Full analysis of these data must await the availability of discharge data.

Henriksen's (1980) F-factor ($\Delta Ca+Mg/\Delta SO_4$) differs between the two catchments. This factor describes the trajectory over time in Henriksen's empirical monograph for freshwater acidfication. Acid treatment at catchment 2 resulted in F=0.1; about 10 % of the sulfate in runoff above background concentrations was accompanied by Ca+Mg (Figure 7). The remaining 90 % was compensated by decrease in bicarbonate, and increase in H^+ and labile Al. Nitrate concentrations were negligible. At catchment 4, however, F=0.6 (Figure 8). Here nitrate is as important as sulfate. The difference in F-factor could be due to different treatment (sulfuric acid only vs. sulfuric plus nitric) or to inherent differences between the 2 catchments. Further study is required; perhaps switching treatments would provide key information.

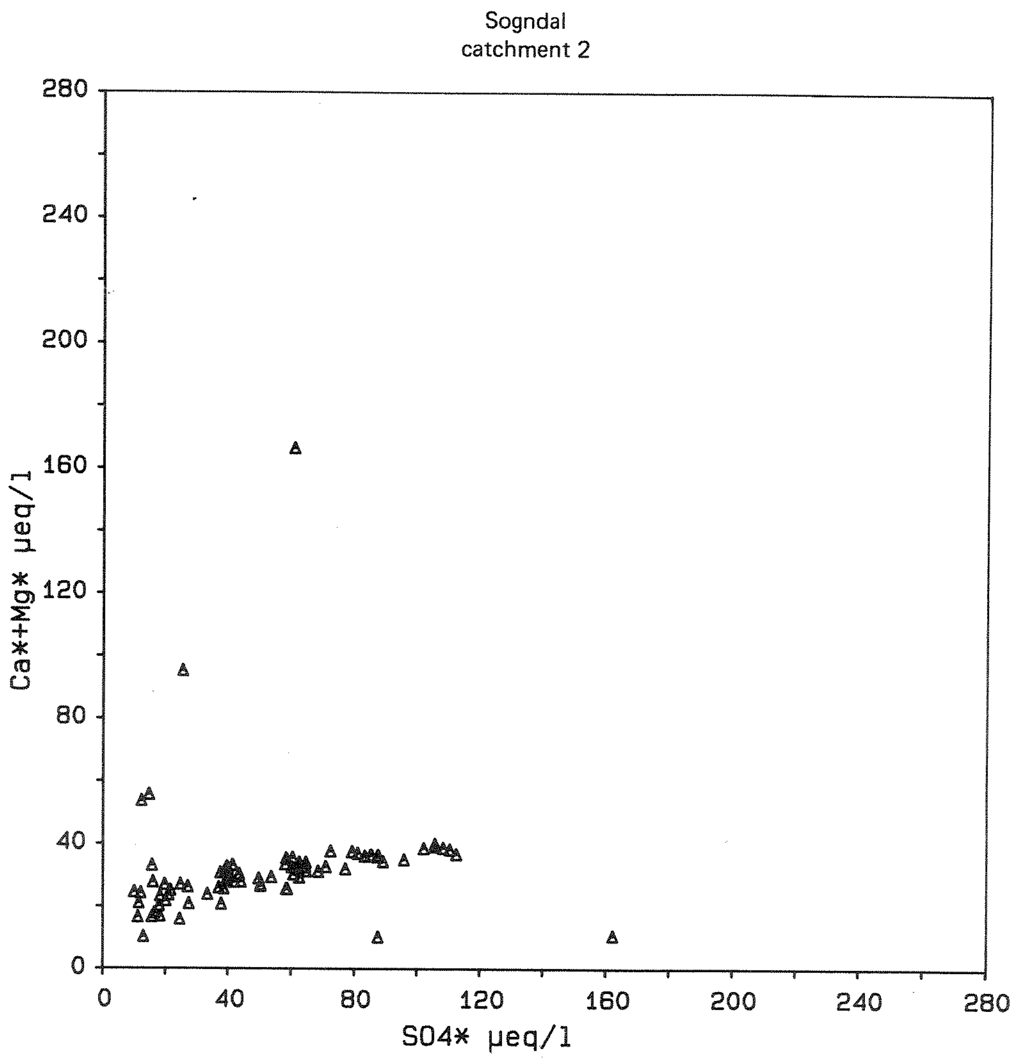


Figure 7. Plot of Ca+Mg concentration against excess-SO_4 levels in runoff from catchment 2 over the period October 1983 - October 1984. Acid treatment resulted in increased SO_4 in runoff, 10 % of which was accompanied by Ca+Mg.

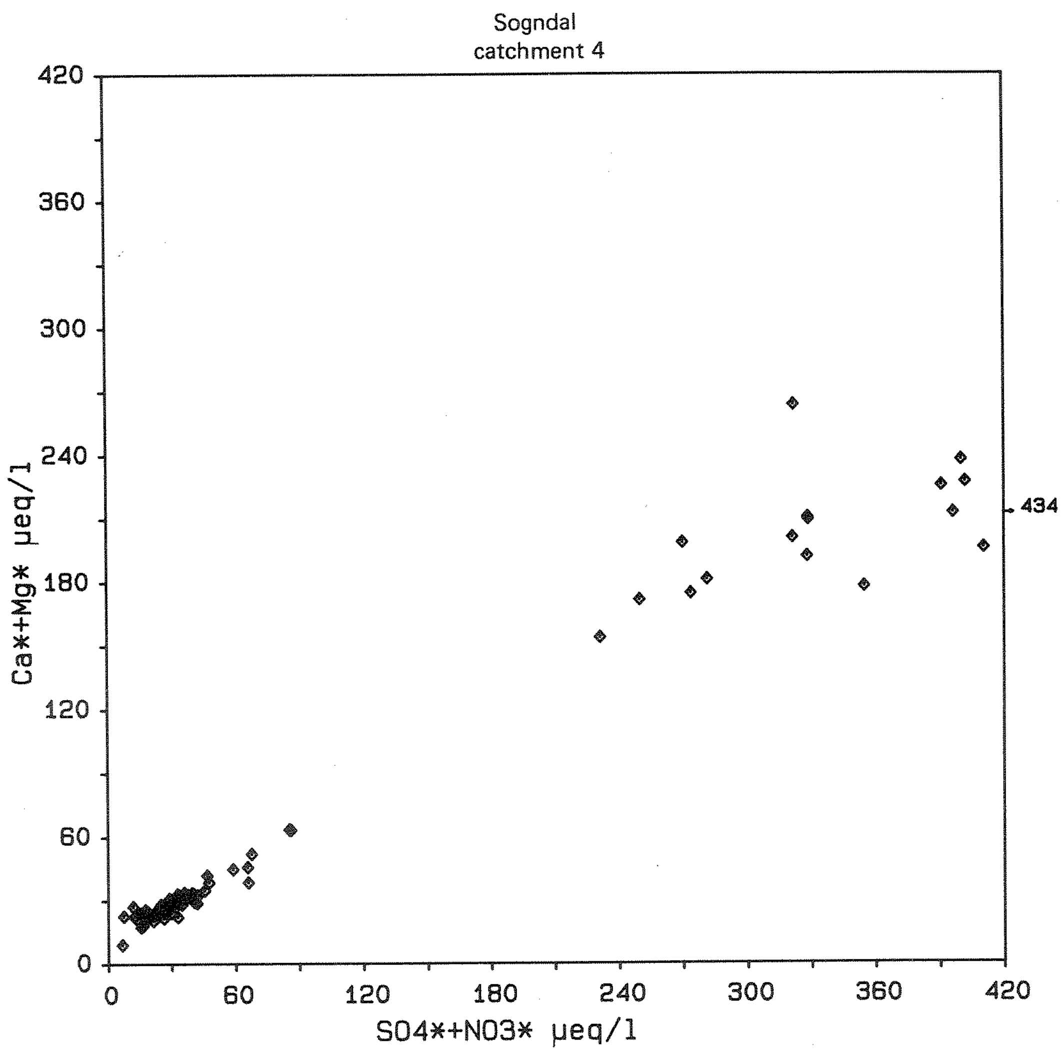


Figure 8. Plot of Ca+Mg concentrations against excess-SO_4 levels in runoff from catchment 4 over the period October 1983 - October 1984. Acid treatment resulted in increased SO_4 and NO_3, 60 % of which was accompanied by Ca+Mg.

5.2. Risdalsheia

Measurements began at Risdalsheia in June 1983 (Table 5). Systematic sampling of runoff began 20 March 1984 at KIM and EGIL catchment and 31 October 1984 at ROLF catchment upon installation of the dams and collection tanks. Microclimate measurements began in August 1984 and have not yet been analyzed. Soil surveys (October 1983, August 1984) and vegetation samples will be reported separately by E. Lotse.

Runoff during snowmelt (20 March - 2 April 1984) was 234 mm at KIM and 189 at EGIL catchment. Spring and summer 1984 was extremely dry; precipitation measured at Birkenes during the period 1 April - 31 August 1984 was 243 mm. Runoff during this period was 18 mm at KIM catchment and 34 mm at EGIL catchment.

Treatment began 16 June 1984 at KIM and 13 June 1984 at EGIL catchment. During the period up to 1 September 71 mm treated rain was applied to KIM and 86 mm acid rain to EGIL catchment. The difference reflects various technical problems during the summer, including lightning damage in July.

The heavy autumn rains typical for southernmost Norway began 7 September. September precipitation totaled 198 mm at Birkenes. At KIM catchment 44 mm was applied; at EGIL 149 mm. The lower amounts were due to continuing technical problems, especially leakage at KIM catchment. The bulk of these problems were attended to during autumn 1984 and as of 1 December 1984 the systems at both catchments were functioning as designed.

The watering systems at KIM and EGIL catchments were closed down for the winter in early December 1984. Both systems had been shut off for several periods during November to avoid frost damage.

With the exception of nitrate the chemical composition of runoff from all three catchments has been similar up to 15 October 1984 (Figure 9). Treatment during the period 15 June - 15 October resulted in significant decrease in nitrate levels runoff at KIM catchment relative to that of EGIL or ROLF catchment (Figure 9). With the exception of runoff following the first major rainfall on 20 September

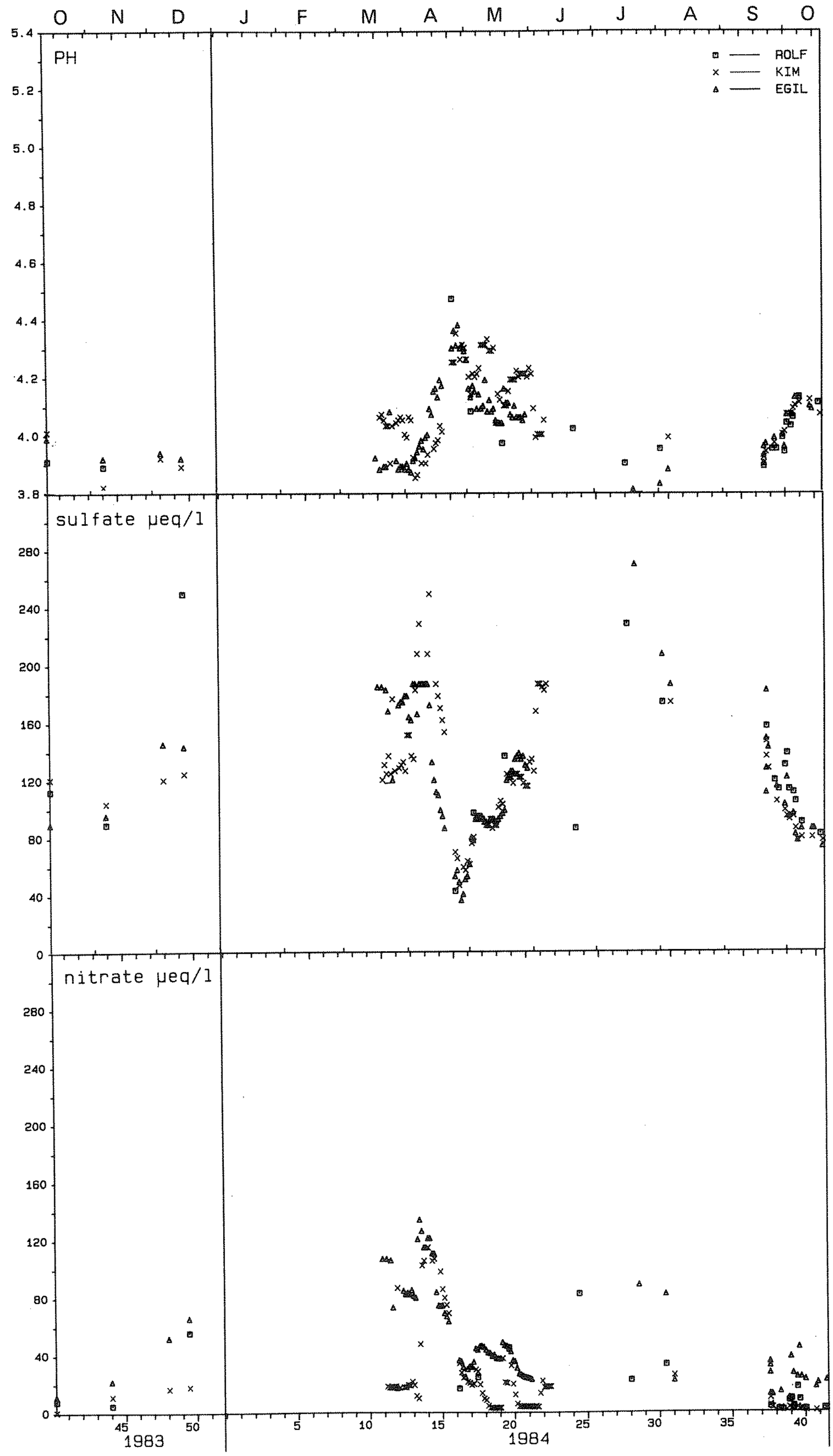


Figure 9. pH and concentrations of sulfate and nitrate in runoff at KIM, EGIL and ROLF catchment at Risdalsheia for the period 15 October 1983 - 15 October 1984.

after prolonged summer drought, nitrate levels at KIM have been below 2 µeq/l, whereas at EGIL and ROLF levels have varied between 5-50 µeq/l. This rapid decrease in nitrate can be ascribed to the treatment; applied rain at KIM no longer contains significant levels of nitrate and ammonia.

5.3. Aluminum species

Three fractions of aluminum are measured routinely in runoff samples from Sogndal and Risdalsheia. These fractions are operationally defined by the analytical procedures (Røgeberg and Henriksen 1984). Total Al is that obtained in samples which have been acidified for several days or more. Reactive Al (total monomeric) (RAL) is that obtained immediately after addition of acid. Non-labile (organic) monomeric Al (NLAL) is that fraction of reactive Al that passes an ion-excharge column. Labile (inorganic) monomeric Al (LAL) is that fraction of reactive Al that is retained by the ion-exchange column. Operationally labile Al is calculated as the difference in reactive Al less non-labile Al.

Aluminium concentrations in runoff at Sogndal and Risdalsheia are a function of pH and concentration of total organic carbon (Table 6). Inorganic Al (LAL) levels increase with decreasing pH (Figure 10). The points from Risdalsheia and Sogndal cluster into two district groups indicating that inorganic Al solubility is controlled by two different solid phases. At Sogndal solibility is higher suggesting control by amorphous aluminium hydroxide, whereas at Risdalsheia a less soluble phase is indicated. This site difference may in turn be related to differences in soil chemistry, in particular soil pH (E. Lotse, in prep.). Organic Al levels (NLAL) increase with increasing concentrations of total organic carbon (Figure 11). At Sogndal concentrations of TOC are low, and thus NLAL levels are low and the linear regression is not significant.

Table 6. Linear regressions of various aluminium fractions on pH and total organic carbon in runoff at Sogndal and Risdalsheia. Units: Al µg/l, H^+ µeq/l, TOC mg C/l.

1.	Inorganic aluminium (labile Al; LAL)				
	Sogndal	log LAL = 8.7 - 1.36 pH		r=0.87	n=241
	Risdalsheia	log LAL = 8.2 - 1.49 pH		r=0.68	n=193
2.	Organic aluminium (non-labile Al; NLAL)				
	Sogndal	not significant			
	Risdalsheia	NLAL = 43.5 + 12.2 TOC		r=0.75	n=193
		NLAL = -13 + 11.8 TOC + 0.7 H^+		r=0.78	n=193
3.	Reactive aluminium (RAL)				
	Sogndal	RAL = -13 + 9.8 TOC + 12.8 H^+		r=0.82	n=241
	Risdalsheia	RAL = -61 + 0.8 TOC + 4.7 H^+		r=0.84	n=193

At Risdalsheia muliple linear regression reveals a small but significant correlation between organic Al and H^+ levels. A possible explanation might be that at higher H^+ levels the concentration of inorganic labile Al are higher and thus favoring greater complexing with organic ligands at a given concentration of TOC. This hypothesis should be tested experimentally.

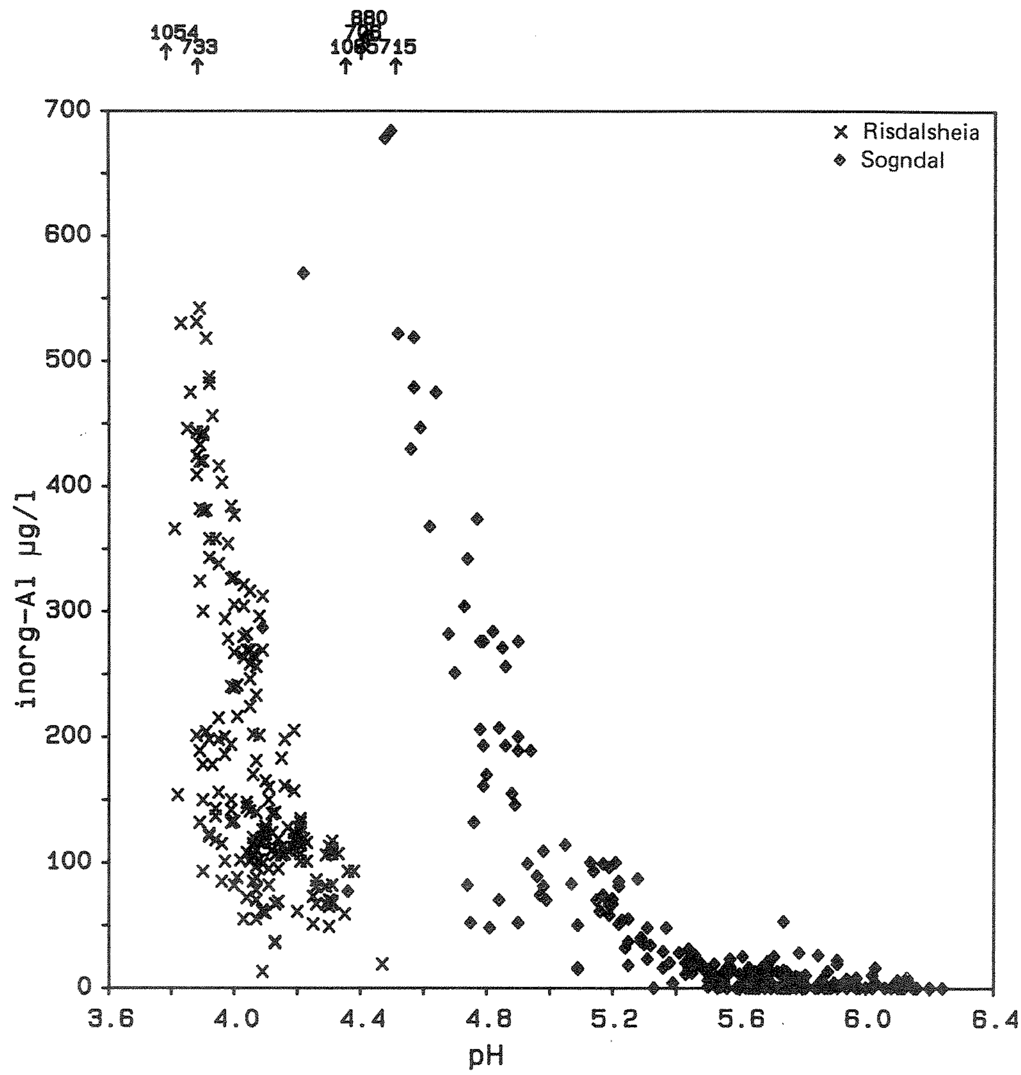


Figure 10. Scatter diagram for pH and concentrations of inorganic-Al (labile Al) in runoff samples collected October 1983 - October 1984 at Sogndal (n=187) and Risdalsheia (n=193).

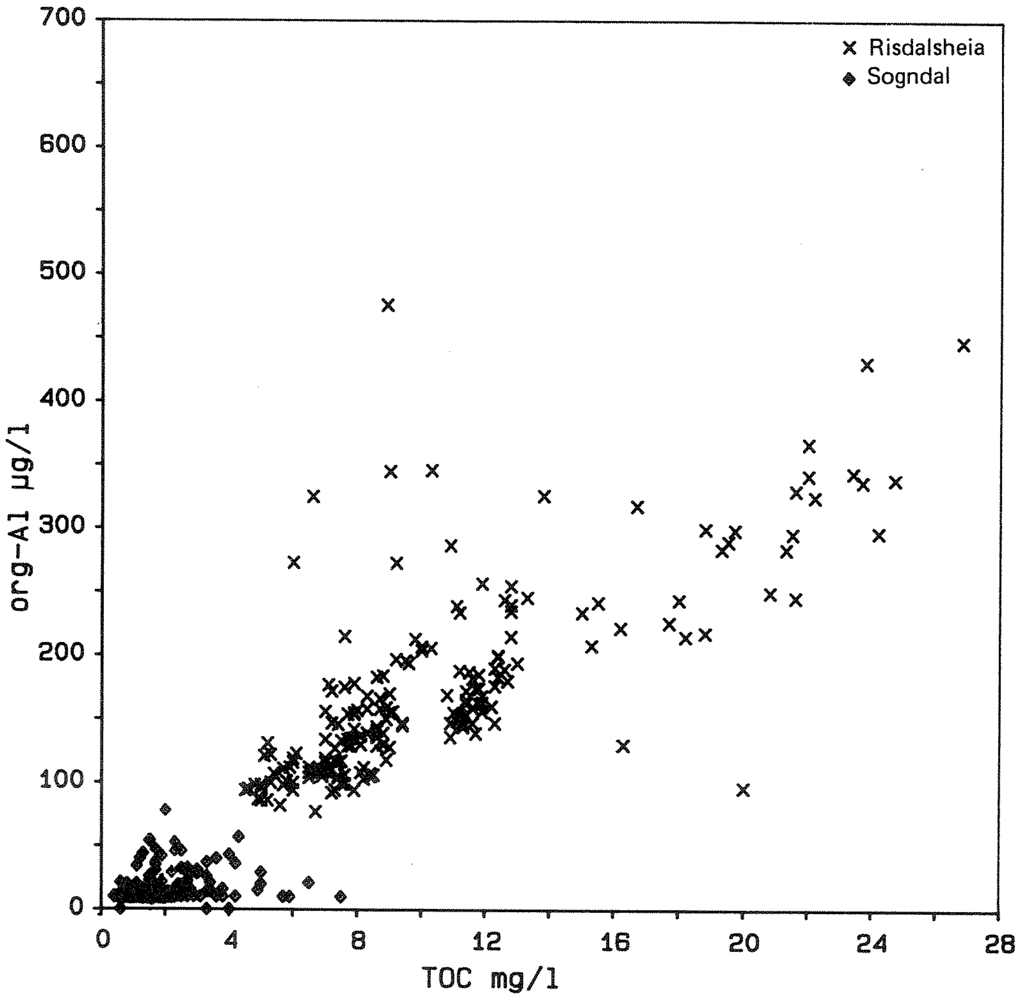


Figure 11. Scatter diagram for TOC and organic-Al (non-labile Al) in runoff samples collected October 1983 - October 1984 at Sogndal (n=187) and Risdalsheia (n=193).

6. SITE VISITS

Project RAIN has received considerable attention both domestically and internationally. In 1984 the project sites were visited by scientists from USA, Canada, Germany, Sweden and Norway and politicians from several countries including 2 delegations from the British House of Commons, the British Parlinentary Under-Secretary of State at the Department of the Environment, and the environment minister from the German Democratic Republic.

7. AUXILIARY PROJECTS

Project RAIN provides an experimental base upon which related studies can build. Several research groups have expressed interest in using the project RAIN sites to study aspects not included in the original design. For example, Prof. Eilif Steinnes, Norwegian Technical University at Trondheim, has obtained independent funds to investigate heavy metal content of plants and soils of the RAIN sites. Other auxiliary research now at the discussion stage includes aluminum mobility in soils, rates of chemical weathering, and aquatic invertebrate organisms at Sogndal.

The project currently has a major proposal under review to the United States Environmental Protection Agency for research on soils, soil solution chemistry, vegetation, and modelling work in conjunction with the RAIN project. A second proposal has been submitted to the Norwegian Hydrological Committee to support hydrological studies at the Risdalsheia site.

8. PLANS FOR 1985

8.1. Technical aspects

The major technical task for 1985 is the production of artificial snow beneath the roofs at Risdalsheia. Commercial snow-making equipment will be used to produce snow 3-4 times during the winter. All other technical aspects have been dealt with in 1983 and 1984. Thus 1985 will be mainly routine maintenance. It is proposed that a reserve fund be established in 1985 to cover repairs and unforeseen technical problems.

8.2. Scientific aspects and reporting

Responsibility for data treatment within project RAIN has been divided among the co-investigators in the project (Figure 3). Richard Wright (NIVA) is responsible for the annual report, description of the project and the sites, water chemistry and hydrology and integration of water chemistry with soil and precipitation chemistry data. Arne Semb (NILU) is responsible for precipitation chemistry. Hans-Martin Seip (SI) is responsible for the effect of the roofs on microclimate. Nils Christophersen (SI) is responsible for hydrochemical models. Erik Lotse (SLU) is responsible for soil and vegetation studies. Reports or publications on several of these topics are anticipated in 1985.

A multi-authored contribution with the title "Project RAIN: experimentally changing the acid deposition to whole catchments" has been submitted to the Muskoka Conference '85.

The preliminary results from 1984 raise several new scientific questions that should be followed up in 1985. A modest expansion of the scientific scope of the project is proposed for 1985. New aspects include:

1. <u>deposition vs. concentration at Sogndal</u>. The 1984 summer treatments resulted in marked pH depressions in runoff to as low as 4.5. Applied rain was pH 3.2. The question arises if depression to this low pH would also occur if the applied rain had a

pH of, say, 3.8. This question is important in that with respect to toxicity to fish and other aquatic organisms it is the concentration of H^+ and labile Al that is critical, whereas with respect to acidification of soils it is the deposition that is of central interest. The problem is one of intensity factor (pH in runoff) vs. capacity factor (acid neutralizing capacity of soils). For 1985 the experiments at Sogndal should be expanded to include treatment of a third catchment with precipitation of lower acidity.

2. Al speciation in runoff. Preliminary results from Sogndal indicate that inorganic labile aluminum is readily mobilized by treatment with acid rain. At Risdalsheia runoff has low pH 3.9-4.2 and preliminary calculations suggest that the labile Al here is primarily in Al^{+3} form. These data coupled with Al-speciation data from other sites in Norway should be combined and analyzed to evaluate the robustness of the analytical methods used (separation by ion-exchange column), the sources of labile Al in runoff, and the effect of pH and organic matter content on Al speciation.

3. Al solubility governed by different solid phases at Sogndal vs. Risdalsheia. Preliminary results suggest that an amorphous aluminium hydroxide phase controls labile Al-concentrations in runoff at Sogndal, whereas at Risdalsheia this phase is appasently absent. These results have implications for (1) short-vs. long-term response to acid deposition, (2) soil chemistry, and (3) reversibility of acidification.

4. Decline in nitrate concentrations in runoff at KIM catchment. The immediate response to decrease in nitrogen loading suggests that the catchment was oversaturated with respect to nitrate prior to treatment. The terrestrial ecosystem was unable to retain all of the incoming nitrogen, perhaps a reflection of changes in the nitrogen cycling due to acid deposition. Nitrate deposition contributes to streamwater acidification at Risdalsheia.

9. REFERENCES

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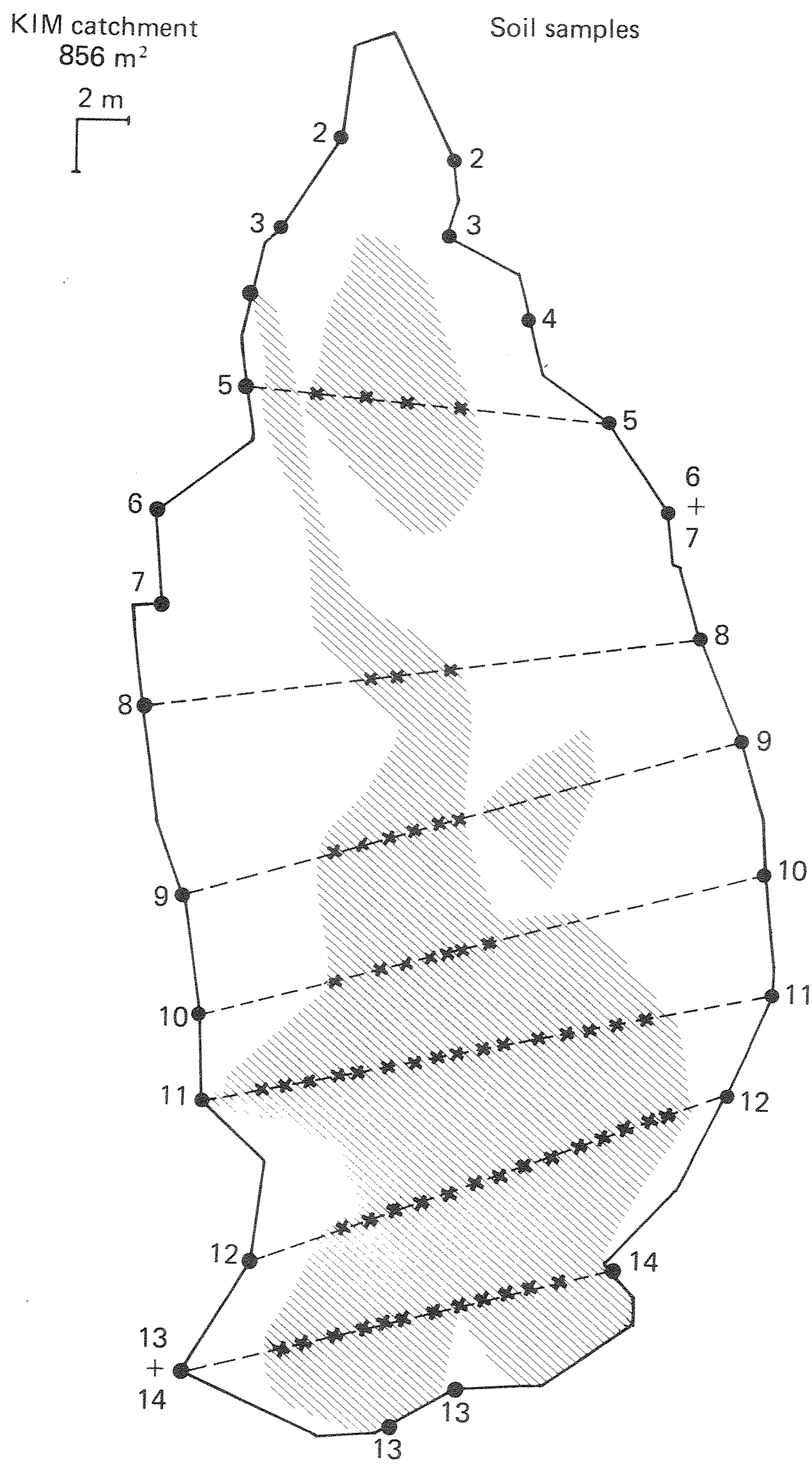
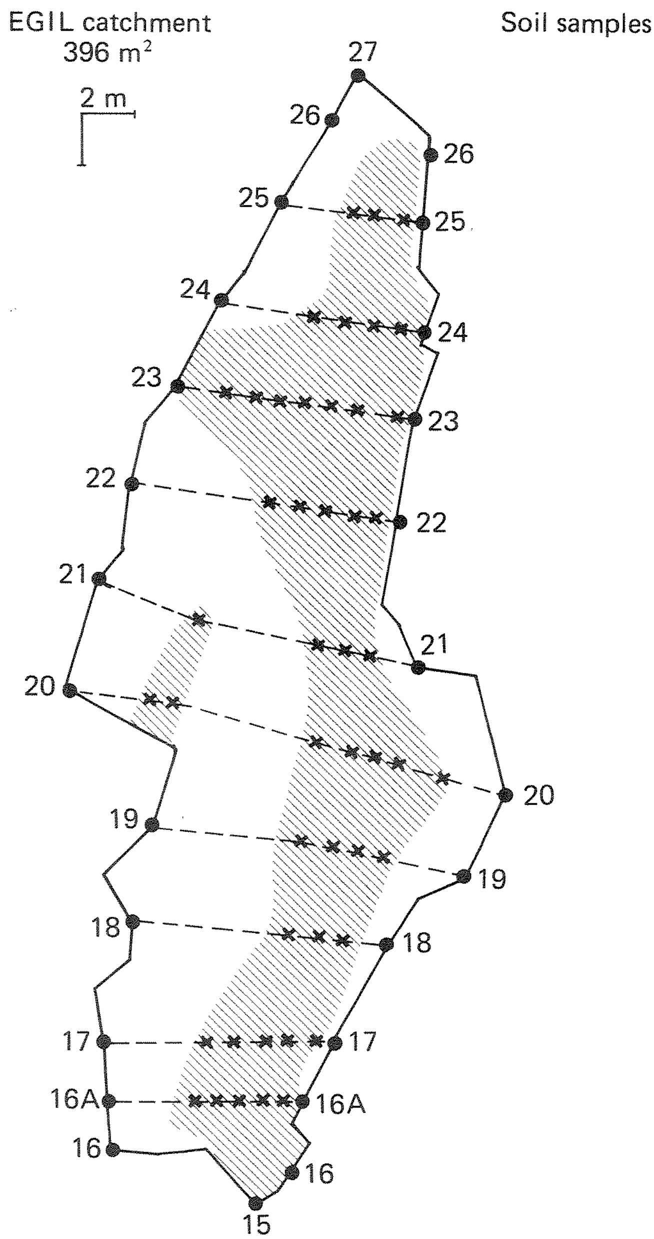
Appendix 1. Maps of the catchments at Risdalsheia showing soil sampling points (hatched areas are soil covered) and areas covered by the roofs.




EGIL catchment
396 m²
Soil samples
2 m
27
26
26
25
25
24
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20
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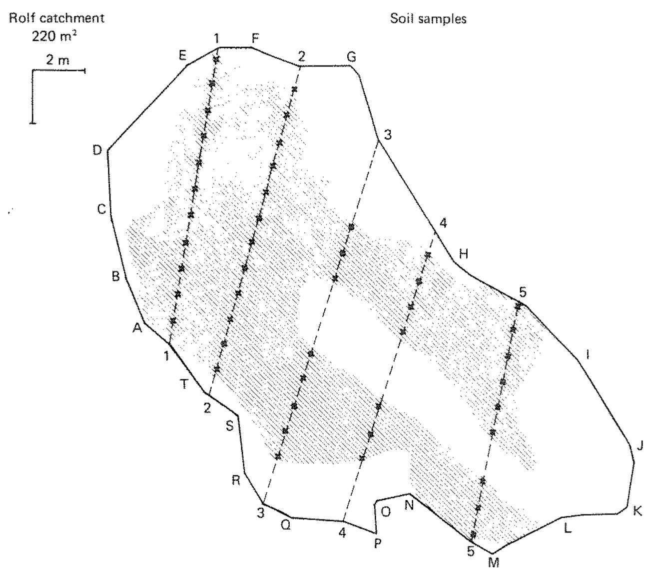
Rolf catchment
220 m²
2 m
Soil samples
A
B
C
D
E
F
G
H
I
J
K
L
M
N
O
P
Q
R
S
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1
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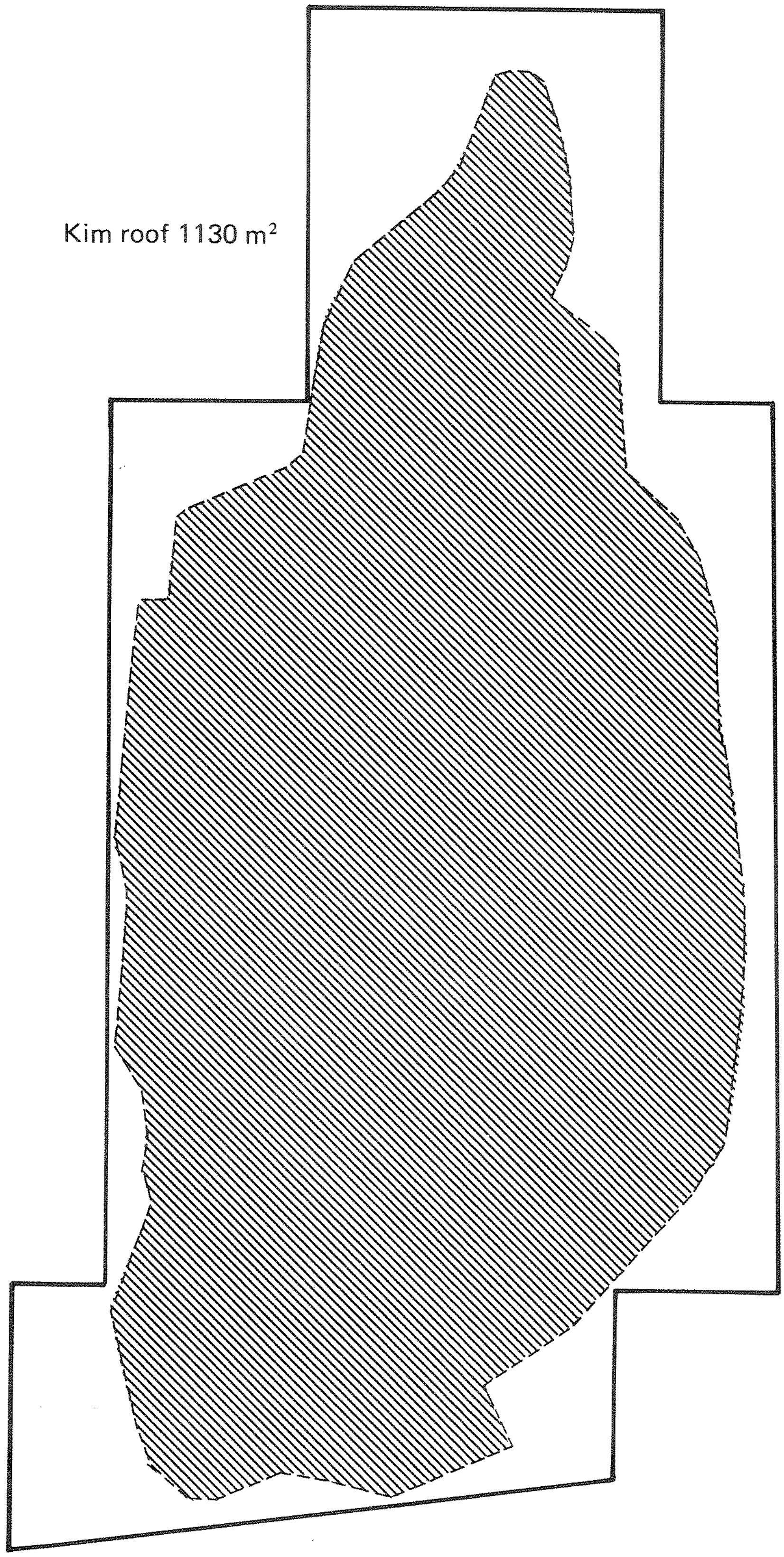
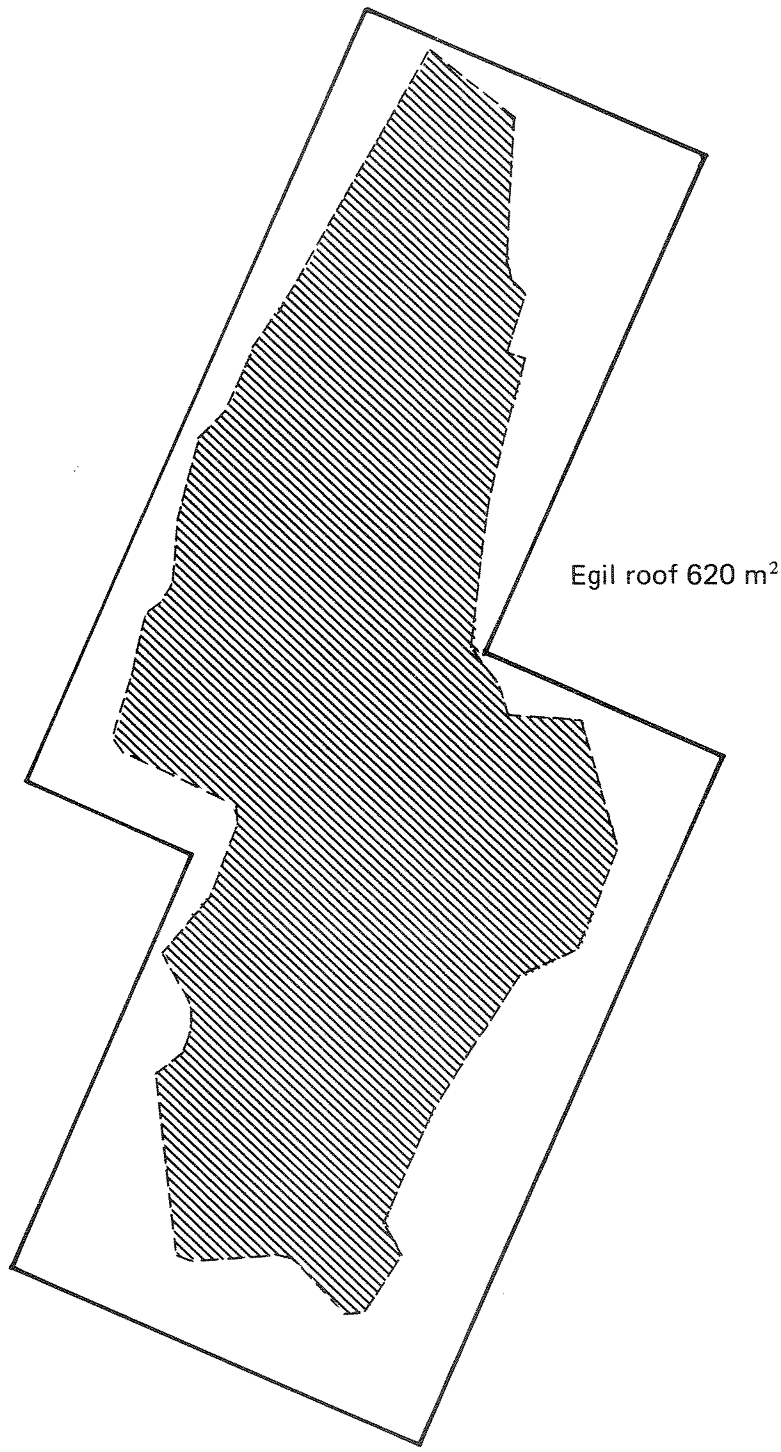
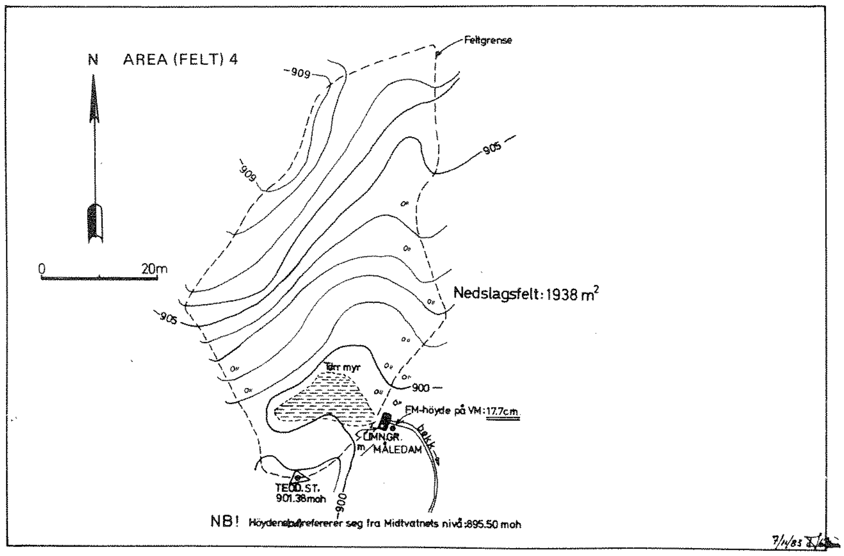
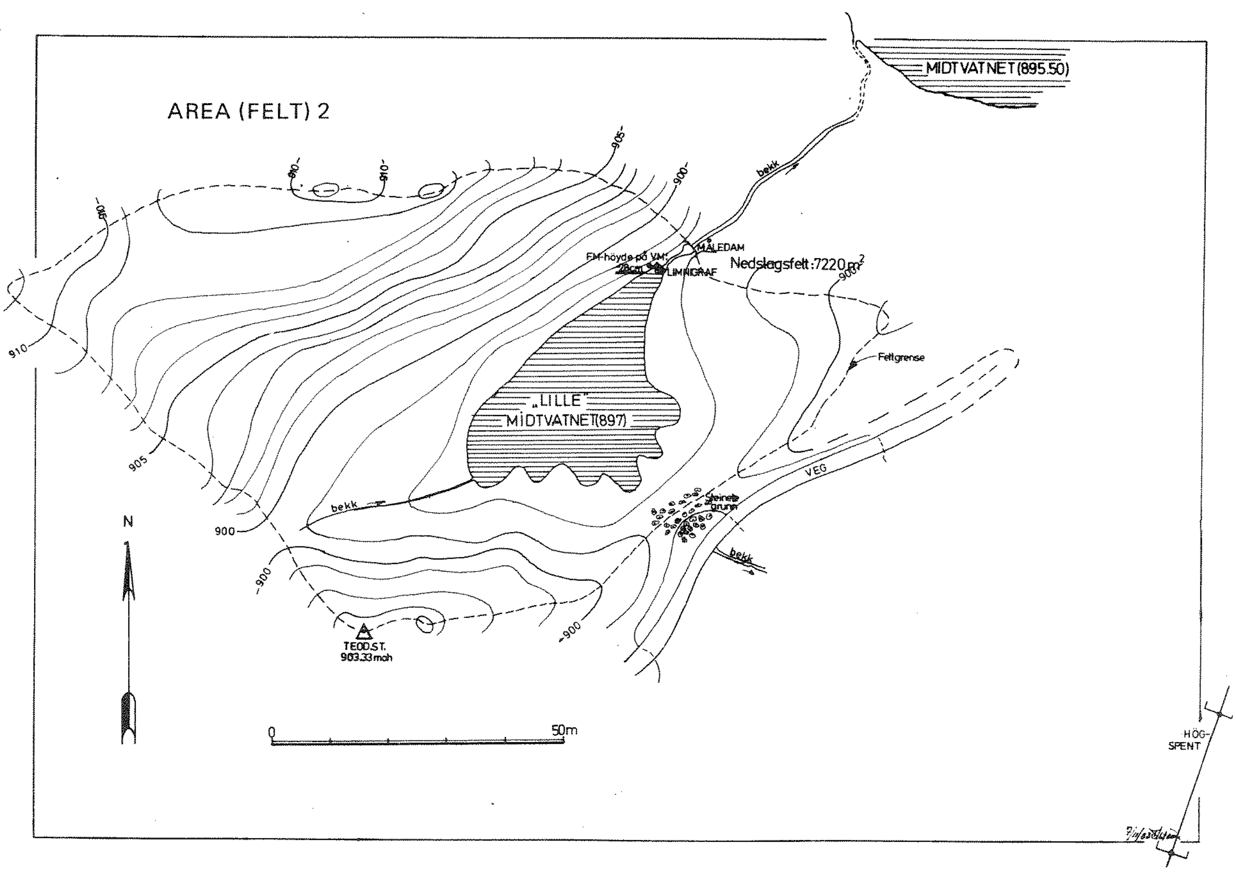
5

Kim roof 1130 m²




Egil roof 620 m²





Appendix 2. Topographic maps of the two treated catchments at Sogndal (courtesy of B. Slettaune, NVE).