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RAIN project. Annual report for 1985

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RAIN PROJECT

Annual report for 1985

Oslo, May 1986

Richard F. Wright
Egil Gjessing

ABSTRACT

Project Rain (Reversing Acidification In Norway) is a 5-year international research project aimed at investigating the effect on water and soil 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.

Treatment at Sogndal commenced April 1984 with the acidification of the snowpack by addition of sulfuric acid (SOG2) and a 1:1 mixture of sulfuric and nitric acids (SOG4). Results from 1985 indicate continued and significant response in runoff chemistry to the acid treatment. Catchment SOG2 did not recover fully during the winter of 1984-85. Most of the added acid continued to be retained in the catchments during 1985. Response during snowmelt 1985 was modest relative to 1984.

At Risdalsheia treatment began in June 1984 with the mounting of the transparent panels on the roofs at KIM catchment (treatment by deacidified rain) and EGIL catchment (control with ambient acid rain). After 1 1/2 years of treatment about 1100 mm of clean precipitation had been applied at KIM. The response in runoff chemistry begun in 1984 continued in 1985; input-output budgets indicate that nitrate is reduced by 75% and sulfate by 35% in runoff from KIM relative to EGIL. Reduction in these anions were balanced by lower levels of most cations.

Aluminum concentrations in runoff at Sogndal indicated a lower solubility of labile Al in 1985 relative to 1984. Solubility with respect to $Al(OH)_3$ expressed as (3 pH - p Al^{+3}) was 7.7 in 1985 and 9.1 in 1984. Acid addition has perhaps depleted a reservior of readily-soluble aluminum in the streambed at these catchments.

The treatments continue in 1986-87. Project RAIN provides experimental evidence bearing on target loading, reversibility of acidification, and the processes linking acid deposition, soil acidification and freshwater acidification.

PREFACE

A large number of individuals and insitutes have cooperated in the RAIN project in 1985. The project scientists include N. Christophersen, E. Lotse, E. Gjessing, H.M. Seip, A. Semb, and R. F. Wright. Technical staff includes S. Andersen, H. Efraimsen, R. Høgberget, A. Rogne, B. Sletaune, R. Storhaug, and K. Wedum. NILU, NIVA, and 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, and A. Risdal provided local assistance.

Financial support in 1985 came from the Norwegian Ministry of Environment, The Royal Norwegian Council for Scientific and Industrial Research, the Ontario Ministry of the Environment, Environment Canada, and the Swedish National Environmental Protection Board.

In 1985 several auxiliary projects were associated with the RAIN project. These include a study of water movement at EGIL catchment funded by the Norwegian Hydrologic Committee (M. Hauhs, NIVA, principal investigator), and two projects financed by the Surface Water Acidification Programme (SWAP) (The Royal Society, the Norwegian Academy of Science and Letters, and the Royal Swedish Academy of Sciences), modelling the acidification process (N. Christophersen, SI, principal investigator) and weathering studies in conjunction with the RAIN project (R. Wright, NIVA, and E. Lotse, SLU, co-principal investigators).

INTRODUCTION

Vigorous efforts to obtain reductions in the emissions of acidifying compounds SO_2 and NOx to the atmosphere are in part based on the premise that such reductions will restore acidified waters. The magnitude and rate of response of natural ecosystems to changes in acid loading is, however, not well known, largely because such effects have been difficult to document in the absence of large-scale reductions. We have now begun large-scale manipulations of natural headwater catchments in Norway. Project RAIN (Reversing Acidification In Norway), a 5-year international research project,

comprises two parallel experiments in which the response of soil and runoff chemistry to changes in loading of strong acids from the atmosphere are studied (Wright 1985, Wright et al. in review). The RAIN project builds on short-term pilot-scale experiments conducted in Norway by Seip et al.(1979) and Christophersen et al. (1982). The project provides information on reversibility of acidification, rate of response and target loadings.

At Sogndal, a "clean" area in western Norway, we are acidifying two pristine catchments by addition of sulfuric (SOG2) and a 1:1 mixture of sulfuric and nitric acids (SOG4), respectively (Figure 1, Table 1). At Risdalheia, an acidified area in southernmost Norway, we have excluded acid precipitation from a small catchment (KIM) by means of a roof and are watering with clean precipitation beneath the roof (Figure 1, Table 1).

Project RAIN is a 5-year study and began in June 1983. The project plan calls for approximately one year of pre-treatment data, 3 years of treatment and 1 year of post-treatment recovery data. The first year was devoted to selection of sites, collection of pre-treatment data, and design, construction and installation of roofs, watering systems, weirs and sampling devices. The scientific program currently includes measurements of precipitation volume and chemical composition, soil chemistry, and runoff volume and chemical composition. The RAIN project design, organization, site descriptions and results obtained through 1984 are described in the annual report for 1984 (Wright 1985). The first year's results were presented at the Muskoka '85 conference (Wright et al. in review). We report here results obtained in 1985.

METHODS NEW IN 1985

Carbon dioxide in soil air. Beginning in May 1985 concentrations of CO_2 in soil air are measured fortnightly during the snow-free season in KIM, EGIL and ROLF catchments. The sampling and measurement procedure entails insertion of thin steel tubes into the soil to a depth of about 30 cm. Three tubes were installed at each site. A metered volume of soil air is withdrawn and passed through a disposable glass column containing silica gel particles impregnated with a hydrazine compound and a redox indicator (crystal violet). The CO_2 concentration is related to the length of penetration of blue color in the glass tube and is mesured in the field. We use tubes supplied by Draeger Inc., Luebeck, West Germany. The method is

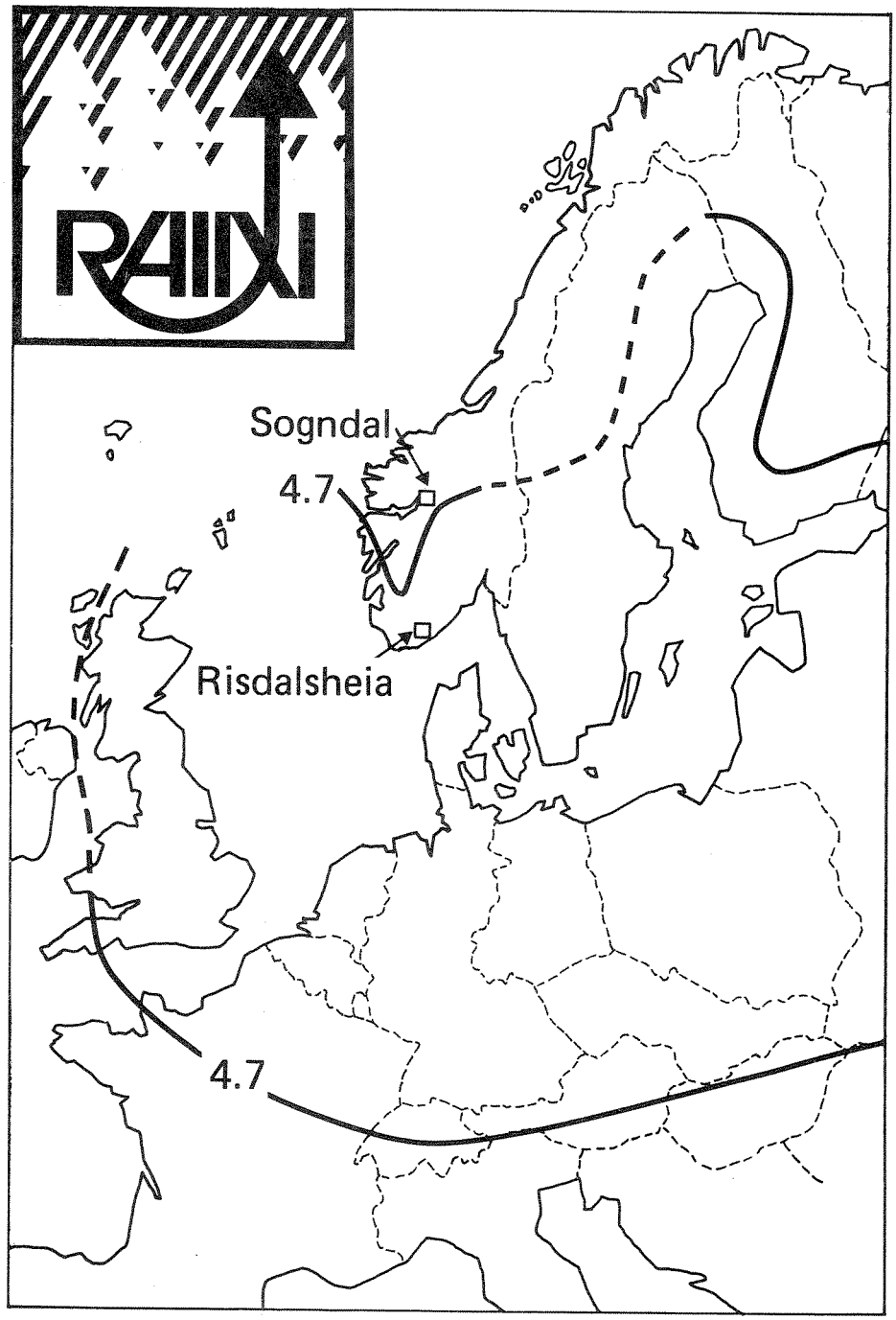


Figure 1. Location of the experimental catchments in project RAIN. Areas within the pH 4.7 isoline receive precipitation with a yearly weighted-average pH below 4.7.

Table 1. RAIN project. Overview of the experimental catchments and treatments.

Sogndal. Acid addition experiments.

Catchment	Treatment	Area
SOG1	control	96300 m^2
SOG2	H_2SO_4	7220 m^2
SOG3	control	43200 m^2
SOG4	$H_2SO_4+HNO_3$	1940 m^2

Risdalsheia. Acid exclusion experiments.

Catchment	Treatment	Area
KIM	roof, clean rain	860 m^2
EGIL	roof, acid rain	400 m^2
ROLF	no roof, acid rain	220 m^2

described by Miotke (1974) and a typical application by Buyanovsky and Wagner (1983).

Carbon dioxide levels in soil air exhibit a seasonal pattern with levels near atmospheric (log pCO_2 -3.5) in spring and autumn with a maximum in mid-to-late summer at 10-50 times atmospheric (log pCO_2 -2.5 to -1.8) (Figure 2). This pattern is typical of forest soils (Miotke 1974, Cosby et al. 1985) and reflects the production of CO_2 from respiration, a temperature-dependent process. These soil CO_2 data are to be used in application of predictive acidification models with the RAIN catchments.

New analytical methods for sulfate and chloride. Beginning with samples collected 13 September 1985 the routine analytical method for sulfate and chloride were changed from automated colorimetry (AC) to ion chromatography (IC). Prior to this change 43 samples from Risdalsheia and 35 samples from Sogndal were analyzed in parallel by both methods. Analysis of precipitation samples at NILU has been by IC throughout the RAIN project.

For sulfate humus coloring interferes with the Ba-thorin method. The parallel analyses revealed no statistically-significant difference in the 35 samples from Sogndal (Figure 3). These have TOC less than 3 mg/l. The samples from Risdalsheia are highly colored (TOC 8-31 mg/l). Here there was a significant difference (Figure 3). Linear regression gives SO_4-IC = -2.2 + 1.31 SO_4-AC with r^2 = 0.88 and $p < 0.001$ for the 43 samples and units of mg/l. The difference (SO_4-IC) - (SO_4-AC) is not significantly correlated to TOC for the 43 Risdalsheia samples.

Under the assumption that the IC gives the correct result, the sulfate data from Risdalsheia from the inception of the RAIN project until 13 September 1985 must be corrected for this systematic analytical error. This correction has been made with the regression equation above. The data reported have been so corrected.

For chloride the parallel analyses indicate no significant difference between the automated thiocyanate method and ion chromatography. Thus no correction is necessary and the change in analytical method for chloride entails no systematic change in reported results.

Pond at SOG2, Sogndal. In November 1985 the bathymetry of the small pond in catchment SOG2 at Sogndal was measured. The pond occupies 800 m^2 (about 11 % of the cathcment area), and its volume of 260 m^3

corresponds to about 36 mm over the entire catchment. This volume is about 2-3 times that applied during each watering treatment (11-15 mm). Mean depth is about 30 cm.

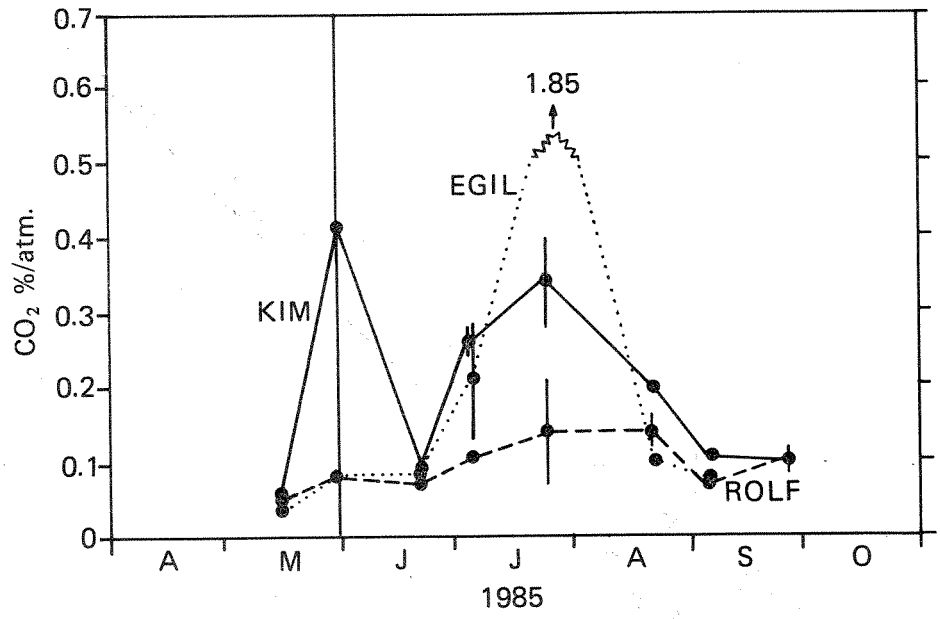


Figure 2. CO_2 content of soil air (mean and standard deviation of 3 samples) at the Risdalsheia catchments during the snow-free season 1985. Atmospheric CO_2 level is 0.035 %.

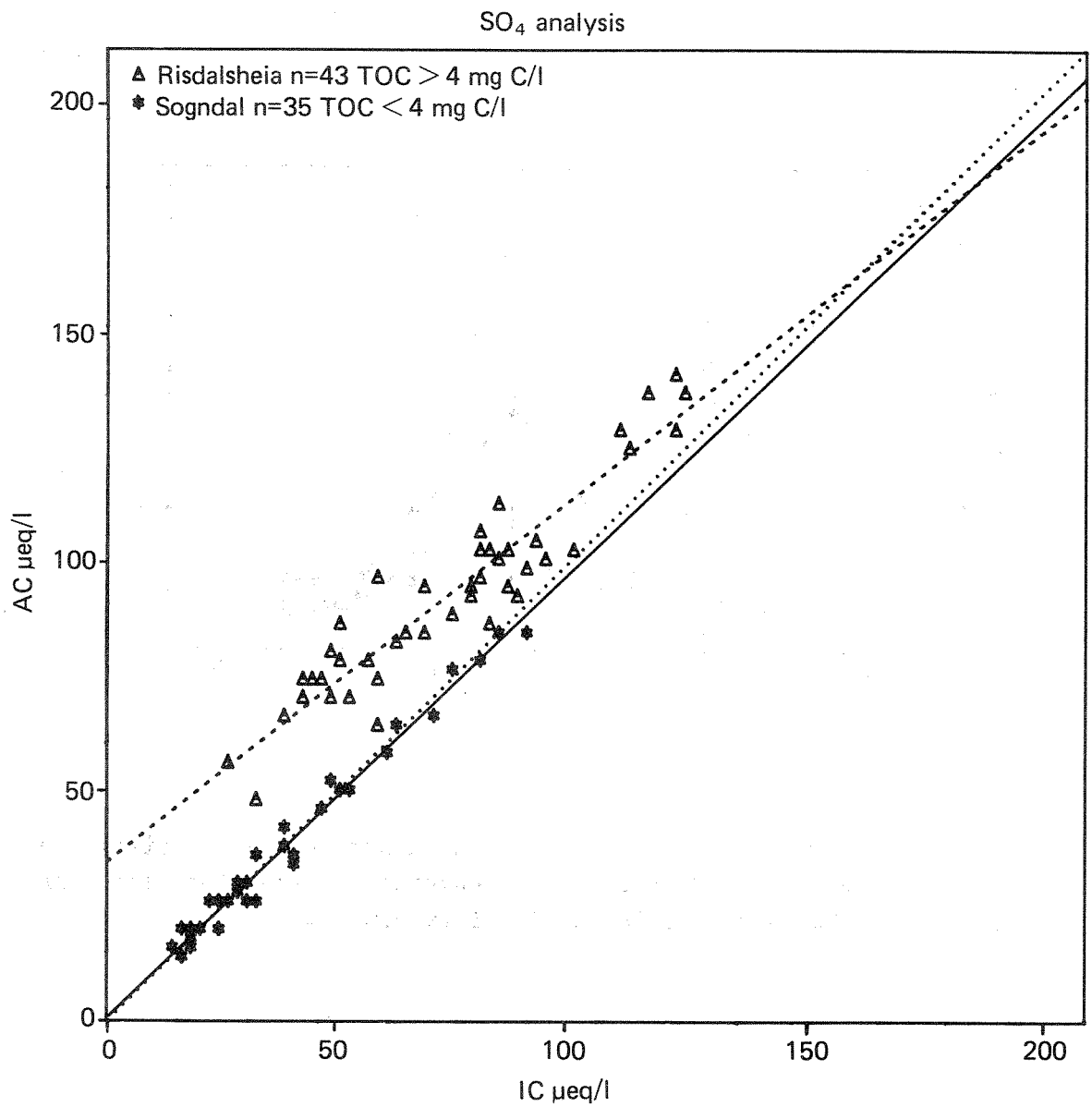


Figure 3. Sulfate concentration determined by automated colorimetry (AC) (thorin method) and by ion chromatography (IC) for samples from Risdalsheia (TOC> 4 mg C/l) and Sogndal (TOC< 4 mg C/l).

New dam at KIM catchment, Risdalsheia. A second dam at KIM catchment was completed on 25 July 1985. Comparison of input and output of water at KIM indicates that about 40% of the runoff had leaked out of the catchment prior to construction of this new dam. The water balance since then indicates that no additional major leaks are present.

Hydrologic measurements at Sogndal. Measurement of both precipitation volume and runoff volume at the Sogndal catchments has been difficult since the start up of these catchments. There are substantial local variations in precipitation especially with elevation. Measurements at Haukås farm at 500 m elevation are not always representative for precipitation at the catchments at 900 m. We thus use the discharge measurements at SOG1 and an estimated evapotranspiration of 100 mm per year to correct the measured precipitation data at Haukås. During winter and spring 1984 the level recorder at SOG1 was damaged by the heavy snowpack and discharge data are thus lacking for this period. The following spring 1985 we measured the snowpack at the four catchments and thereby obtained an estimate of precipitation input which we compare with measured precipitation inputs at Haukås and to runoff during spring snowmelt; these data are used to estimate precipitation input during the previous winter 1984.

The treated catchments SOG2 and SOG4 are both equipped with weir and level recorders. Unfortunately this standard hydrologic equipment is difficult to use at such small catchments (2000-7000 m^2), especially in winter and spring when the entire system freezes. We have thus been forced to estimate discharge from these two catchments during several periods.

We have divided the time since inception of discharge measurements into 5 periods:

period I	830901-831114	(start to freeze up)
period II	831115-840630	(winter and snowmelt)
period III	840701-841112	(summer and autumn)
period IV	841113-850616	(winter and snowmelt)
period V	850617-851101	(summer and autumn)

For each of these periods we have compiled a hydrologic budget for SOG1 and estimated the factor by which precipitation volume at Haukås must be corrected (Table 2). We then take this corrected precipitation volume and estimated evapotranspiration to obtain

Table 2. Hydrologic budgets for the Sogndal catchments. Circled values are estimated. Underlined values are calculated. Unmarked values are measured. SOG3 is assumed to have the same hydrologic budgets as SOG1.

Period	Precipitation measured Haukås		Runoff SOG1 measured		ET est.	Corrected precip.			Added to SOG2 and SOG4		Runoff SOG2					Runoff SOG4				
	mm	Cl meq/m^2	mm	Cl meq/m^2	mm	mm	factor	Cl meq/m^2	mm	Cl meq/m^2	meas. mm	Cl meq/m^2	Corr. mm	factor	Cl meq/m^2	meas. mm	Cl meq/m^2	Corr. mm	factor	Cl meq/m^2
I. 830901-831114	411	19.0	320	10.8	0	320	0.78	14.8	0	0	406	25.1	320	0.79	19.8	600	23.0	320	0.53	12.3
II 831115-840630	493	54.8	656[A]	75.8[A]	0	656	1.35	72.9	0	0	725	56.4	656	0.90	51.0	980	58.5	656	0.67	39.2
III 840701-841112	520	6.9	370	15.5	100	470	0.90	6.2	60	2.2	586	19.6	430	0.73	14.4	350	18.8	430	1.23	23.1
IV 841113-850616	345	13.2	476	14.2	0	476	1.38	18.2	0	0	(570)	(14.0)	476	0.84	11.7	925	20.6	476	0.51	10.6
V 850617-851101	342	11.3	352	9.6	100	452	1.32	14.9	63	1.7	394	10.8	415	1.05	11.4	275	7.6	415	1.51	11.5

[A] Measured flow not available due to damage to level recorder during spring snowmelt. Volume estimated as measured precipitation x1.33. The factor 1.33 comes from the measured runoff/measured precipitation winter 1985 (period IV). Cl flux is scaled to 656 mm runoff.

Table 3. Summary of Sogndal treatments. Catchment SOG2.

Date	Pre-treatment Time	mm	Acid application Time	mm	Post-treatment Time	mm	Acid dose meq/m^2 H^+	SO_4	NO_3
840401			to snowpack	0,01			25.0	25.0	0
840828	1200-1350	3.9	1353-1835	11.0	1840-1910	1.0	11.2	11.2	0
840912	1420-1520	2.2	1520-2005	9.8	2005-2045	1.7	11.2	11.2	0
840926	0720-0820	2.2	0820-1310	11.0	1320-1430	1.9	11.2	11.2	0
841002	0715-0810	2.2	0810-1255	11.0	1255-1350	2.2	11.2	11.2	0
(total added in 1984 meq/m^2: H^+ 69.8, 3.2 Na, 0.2 K, 1.0 Ca, 0.7 Mg, 3.2 Cl, 69.8 SO_4; 60 mm H_2O)									
850329			to snowpack	0.01			25.0	25.0	0
850611	1335-1405	1.1	1410-1900	11.0	1930-2035	2.1	11.2	11.2	0
850821	0800-0900	2.2	0915-1415	11.0	none		11.2	11.2	0
850821	none		1630-2100	11.0	none		11.2	11.2	0
850828	1030-1125	2.2	1125-1655	11.0	1655-1735	1.9	11.2	11.2	0
851001	none		0645-1145	11.0	none		11.2	11.2	0
(total added in 1985 meq/m^2: H^+ 82.6, 2.4 Na, 0.1 K, 0.7 Ca, 0.6 Mg, 1.7 Cl, 82.6 SO_4; 61 mm H_2O)									

Catchment SOG4.

Date	Pre-treatment Time	mm	Acid application Time	mm	Post-treatment Time	mm	Acid dose meq/m^2 H^+	SO_4	NO_3
840402			to snowpack	0.01			25.0	11.2	11.2
840829	0730-0840	2.6	0840-1325	11.2	1325-1400	0.5	11.2	5.6	5.6
840912	0730-0830	2.6	0830-1310	9.6	1315-1350	1.4	11.2	5.6	5.6
840926	1405-1450	1.8	1450-1935	11.2	1935-2010	1.8	11.2	5.6	5.6
841002	1400-1445	2.0	1445-1930	11.1	1930-2010	2.0	11.2	5.6	5.6
(Total added in 1984 meq/m^2: H^+ 69.8, 3.1 Na, 0.2 K, 1.0 Ca, 0.7 Mg, 3.1 Cl, 33.6 SO_4, 22.4 NO_3; 58 mm H_2O)									
850329			to snowpack	0.01			25.0	11.2	11.2
850612	0815-0855	1.8	0930-1400	10.8	1410-1435	1.7	11.2	5.6	5.6
850822	none		0950-1450	10.8	1450-1550	2.0	11.2	5.6	5.6
850823	none		0715-1115	10.8	1115-1215	2.0	11.2	5.6	5.6
850827	1400-1500	1.8	1505-1930	10.8	1930-2030	1.8	11.2	5.6	5.6
851001	none		1235-1735	10.8	none		11.2	5.6	5.6
(Total added in 1985 meq/m^2: H^+ 82.6, 2.5 Na, p.1 K, 0.7 Ca, 0.6 Mg, 1.8 Cl, 40.0 SO_4, 39.7 NO_3; 65 mm H_2O)									

estimated runoff volume at SOG2 and SOG4 for each period. These calculations are summarized in Table 2. The chloride flux as measured and estimated by this procedure is also shown for comparison.

Table 4. Risdalsheia. Major changes in operation

Date	Operation
23 March 1984	Runoff volume measurements begin at KIM and EGIL
13 June 1984	Treatment begins at KIM and EGIL
1 August 1984	New dam at EGIL; all runoff now collected
31 October 1984	Runoff measurements begin at ROLF
19 December 1984	Sprinkler systems closed for winter at EGIL and KIM
29 April 1985	Sprinkler systems opened again at EGIL and KIM
27 July 1985	New dam at KIM; all runoff now collected
15 November 1985	Sprinkler systems closed for winter at EGIL and KIM

TREATMENTS

Sogndal. At Sogndal treatments continued in 1985 using the same procedures as in 1984 (Wright 1985). The acid dose in 1985 was increased by 13%; there were 5 episodes during the summer of 1985 (Table 3). A double episode in August was carried out in conjunction with the MOBILLAB measurements.

During the period 21 August - 1 September 1985 NIVAs newly-developed, continuous water quality monitoring system MOBILLAB was in use at catchment SOG2. MOBILLAB conducts continuous measurement of pH, conductivity, reactive aluminum and non-labile aluminum. A double episode of acid was conducted during this period and MOBILLAB monitored water quality at the outlet of SOG2. In conjunction with these chemistry measurements, fish and insects were placed in cages at the outlets of SOG2 and SOG1 (control), and the mortality was followed. All the fish (brown trout 0+) died within three days at SOG2 (none died in the control). These results will be reported in detail elsewhere.

Risdalsheia. Treatments continued at Risdalsheia in 1985 by the same procedures as in 1984. A summary of major changes in operations is given in Table 4.

RESULTS

Sogndal

At Sogndal both catchments continued to respond immediately and dramatically to the acid treatments in 1985 (Figure 4). Catchment SOG2 did not recover fully to background levels of sulfate during the winter of 1984-85. Sulfate levels during 1985 were consistently higher than those at SOG3, the untreated control catchment. Several of the acid additions in 1985 coincided with major precipitation events, and thus the peaks in sulfate, nitrate, labile aluminum and H^+ were not as pronounced in 1985 as compared with the previous year.

Snowmelt in 1985 again resulted in acidification of runoff at SOG2 and SOG4 relative to the reference SOG3 (Figure 4). SOG2 apparently did not fully recover from the acid treatments the previous autumn and exhibited slightly lower pH levels and higher sulfate levels in baseflow throughout the winter. Minimum measured pH in runoff from SOG2 during snowmelt in 1985 was about 5.0, substantialy higher than the pH 4.1 measured in the first snowmelt of 1984.

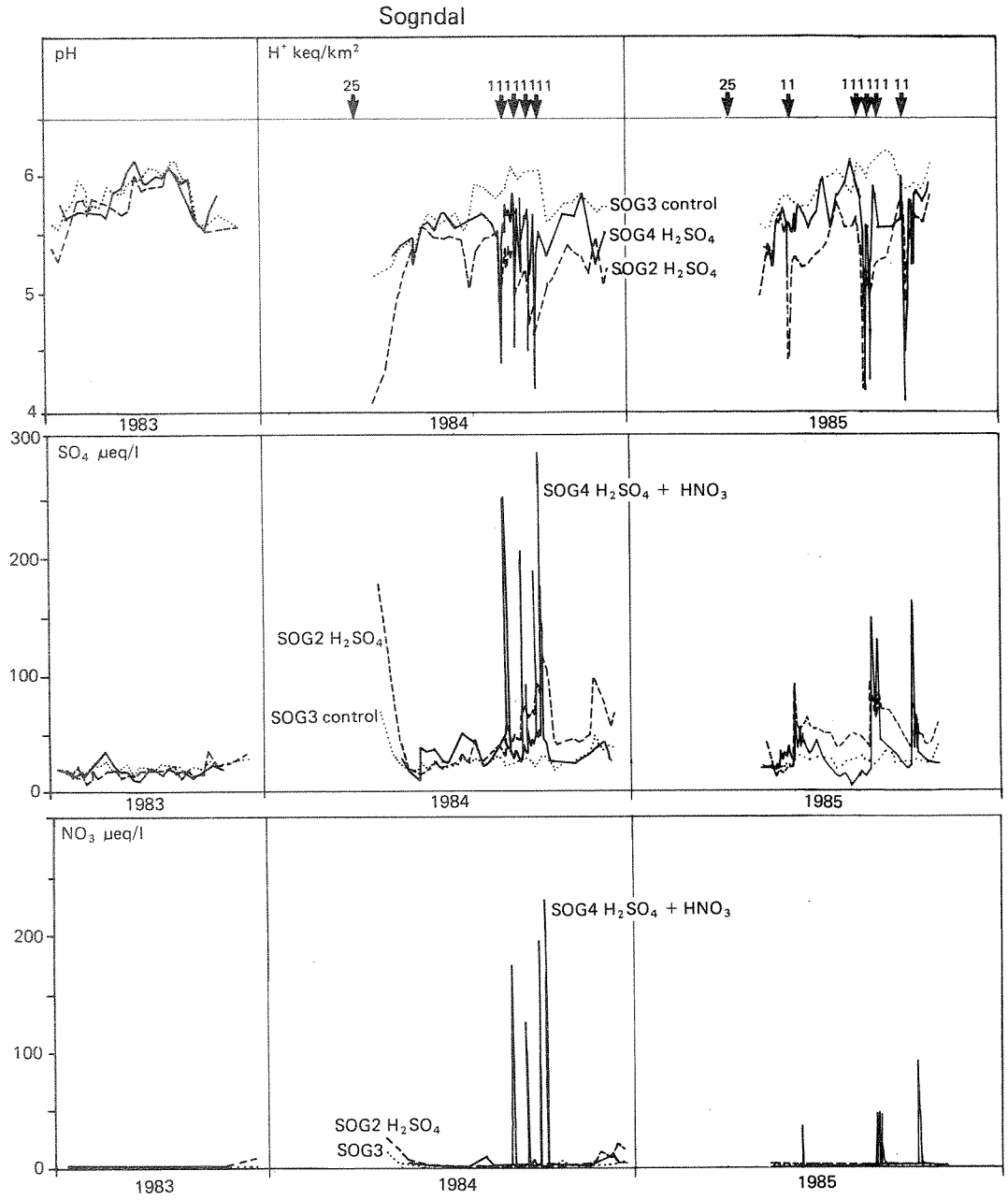


Figure 4. pH and concentrations of sulfate and nitrate in runoff from catchments SOG2, SOG3 and SOG4 at Sogndal over the period June 1983 - June 1985. Treatments began in April 1984 with the acid addition to the snowpack. Acid additions are indicated by arrows.

The first spraying of acid onto the soil in 1985 was carried out in June, only a few weeks following the snowmelt. The response at both catchments SOG2 and SOG4 was similar to that observed following application of acid the previous summer and autumn, except that while the pH levels again dropped to about pH 4.4, the concentrations of the acid anions sulfate and nitrate were considerably lower than at similar pH levels the previous autumn. The runoff in June 1985 was considerably more dilute and contained lower concentrations of the base cations, probably due to the flushing of the soil by large volumes of dilute snowmelt water only a few weeks previously.

Both the treated catchments continued to retain a substantial portion of the added acid (Table 5). At SOG2 about 20% of the added sulfate left the catchment in runoff in 1984 and about 15% in 1985. The difference is probably due to differences in hydrology between the two years. At SOG4 the added nitrate is almost entirely retained in the catchment, although concentrations in runoff reach 200 µeq/l during acid treatments. Nitrate is retained to a higher degree relative to sulfate at SOG4.

Risdalsheia

By 13 November 1985 after 1 1/2 years of treatment a total of 1100 mm of rain and snow have been ion-exchanged and applied beneath the roof at KIM catchment. At EGIL catchment 1350 mm have been applied. During this same period natural precipitation amounted to about 2270 mm. Average-annual precipitation at Birkenes is 1350 mm which is also our best estimate for Risdalsheia.

Thus although treatment has proceeded for 1 1/2 calendar years, the equivalent of only about 1 year of precipitation has been applied to KIM and EGIL. The difference is due to periods with technical problems during which all incoming precipitation is not collected, periods of high precipitation intensity during which the designed capacity of the sprinkler system is exceeded, and the much larger natural snowfall winter 1985 (550 mm) as compared with the 115 mm of artificial snow produced.

Table 5. Input-output budgets for water and major ions at the Sogndal catchments for the periods 831115-841113 (1984) and 841114-851101 (1985). Units: H_2O in mm; ions in meq/m^2/yr.; TOC in mg C/m^2/yr; SiO_2 in mg SiO_2/m^2/yr.

- 1984 -

	SOG1 control		SOG2 H_2SO_4		SOG3 control		SOG4 H_2SO_4 + HNO_3	
	In	Out	In	Out	In	Out	In	Out
H_2O	1126	1026	1186	1086	1126	1026	1186	1086
H^+	19	2	89	9	19	3	89	4
Na	70	90	73	64	70	89	73	55
K	6	11	6	4	6	2	6	2
Na	9	21	10	24	9	22	10	28
Mg	16	15	16	14	16	18	16	12
Al	-	1	-	3	-	1	-	2
NH_4	9	6	9	3	9	1	9	1
NO_3	9	2	9	4	9	2	44	4
Cl^-	79	91	82	65	79	85	82	62
SO_4	30	29	100	43	30	25	65	29
HCO_3	0	15	0	5	0	2	0	7
Σ^+	129	146	203	121	129	137	203	104
Σ^-	119	138	191	113	119	114	192	102
TOC	-	4.4	-	2.7	-	0.8	-	2.9
SiO_2	-	-	-	-	-	-	-	-

Table 5. (cont.)

- 1985 -

	SOG1 control		SOG2 H_2SO_4		SOG3 control		SOG4 H_2SO_4 + HNO_3	
	In	Out	In	Out	In	Out	In	Out
H_2O	928	828	991	891	928	828	991	891
H^+	19	2	102	4	19	2	102	3
Na	41	29	43	28	41	27	43	27
K	6	2	6	2	6	1	7	2
Ca	5	13	5	20	5	13	6	18
Mg	5	7	5	9	5	7	5	8
Al	-	0	-	2	-	0	-	1
NH_4	8	4	8	0	8	1	8	1
NO_3	10	1	10	1	10	1	51	1
Cl	33	24	35	23	33	20	31	22
SO_4	21	18	104	35	21	18	63	20
HCO_3	0	11	0	2	0	5	0	5
Σ^+	84	58	169	65	84	51	170	61
Σ^-	64	54	149	61	64	44	144	49
TOC	-	1.3	-	1.4	-	0.8	-	2.5
SiO_2	-	0.8	-	1.2	-	0.8	-	1.6

The trends in runoff water quality observed in late 1984 at KIM catchment continued in 1985. Nitrate levels remained significantly lower in runoff from KIM than that from both EGIL and ROLF (Figure 5). Sulfate levels from KIM continued at about 60-80 µeq/l whereas runoff from EGIL catchment contained significantly higher levels (90-140 µeq/l).

Sulfate levels in runoff from EGIL continued to exhibit seasonal variations related to deposition and accumulation in the catchment (Figure 5). At KIM, however, the sulfate maxima no longer occur. Concentrations were relatively constant during 1985. Desorption of sulfate from the soils is a process by which sulfate levels in runoff could be kept relatively constant at concentrations higher than inputs.

Input-output budgets for KIM and EGIL catchments illustrate the effects of reduced deposition at KIM. Estimation of dry deposition poses the greatest difficulty in compiling such budgets. For EGIL and KIM catchments we estimate dry deposit during the 1 1/2 years of treatment on the basis of (1) detailed studies of dry deposition at Gårdsjön in Sweden (Grennfelt et al. 1985), and (2) measurements of aerosols and gases at Birkenes (SFT 1985).

We start by assuming that chloride balances in EGIL such that dry deposit of Cl = Cl out - Cl wet in. Next we assume that this Cl comes as marine aerosol with the same chemical composition as seawater and thereby obtain estimates for dry deposit of Na, K, Ca, Mg, and marine SO_4.

Dry deposit of nitrate occurs mainly by gaseous deposition. In 1984 the mean concentration of NO_2 at Birkenes was 1.1 $\mu gN/m^3$ (SFT 1985), which when combined with the deposition velocity for 50% forest and 50% bare ground recommended by Grennfelt et al. (1985) gives 18 meq/m^2 NO_3 for the 1 1/2 year period.

Dry deposit of SO_4 occurs as marine particles, non-marine particles, and SO_2 gas. In 1984 the mean concentration of SO_4 particles at Birkenes was 1.1 $\mu gS/m^3$ (SFT 1985), which when combined with deposition velocity of 0.4 cm/s gives 12 meq/m^2 for the 1 1/2 year

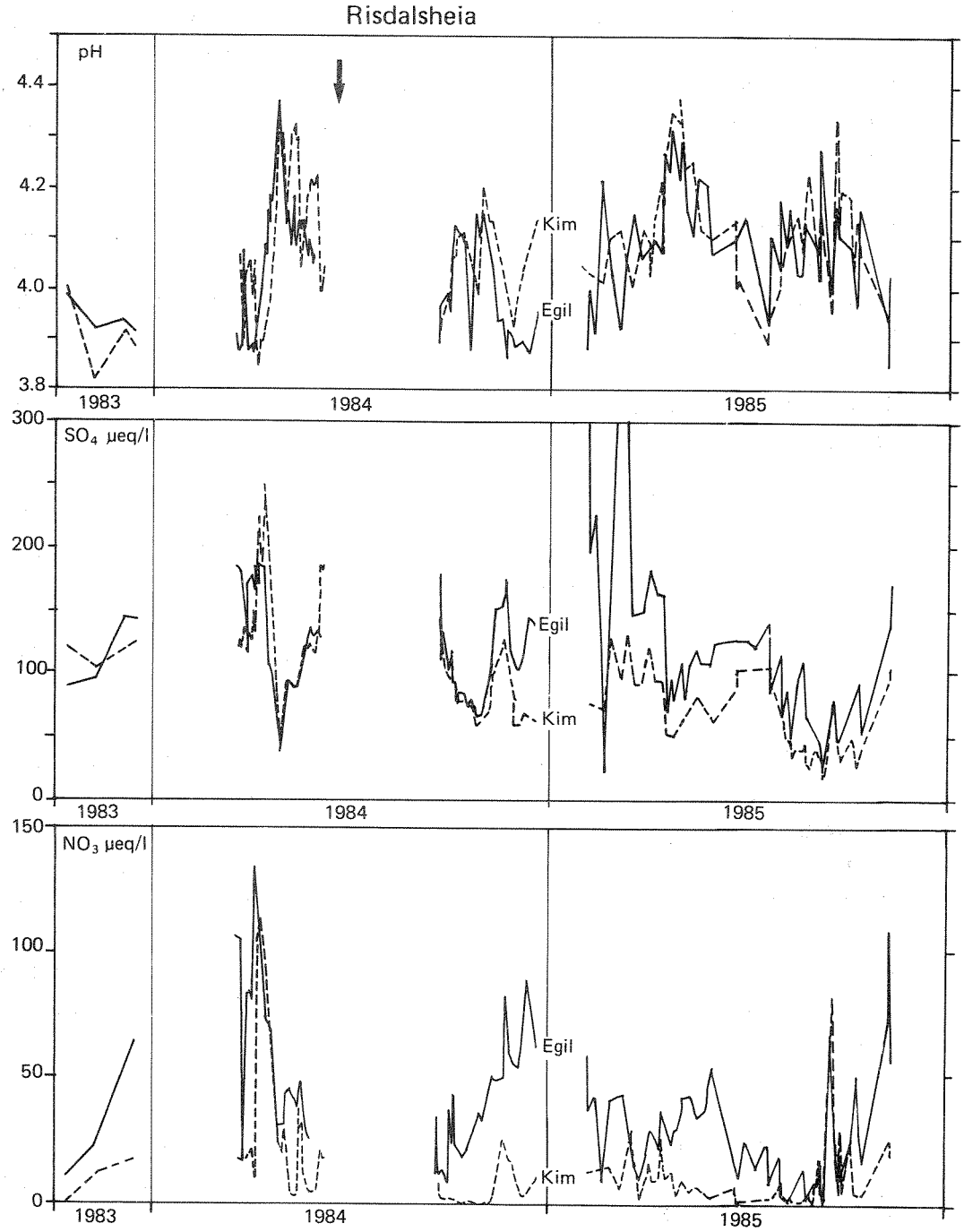


Figure 5. pH and concentrations of sulfate and nitrate in runoff from KIM and EGIL catchments at Risdalsheia over the period October 1983 - June 1985. Treatment began in mid-June 1984 (arrow).

period. Of these about 4 meq/m^2 are marine (from the Cl balance) and 8 meq/m^2 non-marine. For SO_2 gas the mean concentration was 0.7 µg S/m^3 which with a deposition velocity of 0.7 cm/s gives 13 meq/m^2 for the 1 1/2 year period. These deposition velocities are in the middle of the ranges used by Grennfelt et al. (1985) and SFT (1985).

Dry deposit of NH_4 occurs primarily as non-marine particles associated with sulfate. Assuming a molar ratio of 1.5 (Grennfelt et al. 1985) and the non-marine particle deposition of SO_4 of 8 meq/m^2, deposition of NH_4 for the 1 1/2 year period amounts to 6 meq/m^2.

We now obtain an estimate of dry deposit of H^+ by difference from the ionic balance.

H^+ dry deposit = SO_4 dry deposit (non-marine particles plus SO_2 gas) + NO_3 dry deposit (NO_2 gas) - NH_4 dry deposit (non-marine particles)

= 8 + 13 + 18 - 6 = 33 meq/m^2 for the 1 1/2 year period.

For KIM catchment we assume that dry deposit is the same as EGIL for all components. This is probably an overestimate as some of the dry deposition occurs on the roof surfaces. At EGIL catchment this material is recycled and applied in rain beneath the roof in the summer, whereas at KIM this material is removed by the ion-exchangers.

With respect to output budgets we calculate fluxes of Al from measured concentrations of labile inorganic aluminum assuming a valence of +3. Organic anion concnetrations are estimated to make up the ionic balance.

The major features of input-outputs budgets for EGIL catchment are (1) approximate balance of H+ and base cations, (2) net output of Al, (3) large net uptake of NH_4 and NO_3, and (4) approximate balance of SO_4 (Table 6). Cl was assumed to balance. With the exception of the input-output balance for H+ and base cations these results are similar to those generally found at small catchments receiving acid precipitation in Norway (Wright and Johannessen 1980). That base cation outputs equal inputs is due to the extremely thin soils and the large fraction of the catchment at EGIL and KIM with exposed bedrock. Weathering rates are thus low.

Table 6a. Input-output budgets for water and major ions at EGIL and KIM catchments for the $1^1/_2$ year treatment period 13 June 1984 - 14 November 1985. Units: meq/m^2. See text for details.

	EGIL						KIM			
	Input					Out	Input			Out
	Wet	Dry			Total		Wet	Dry	Total	
		marine part.	Acid part.	gases						
H_2O (mm)	1346					1092	1102			835
H^+	64	0	2	31	97	102	10	33	43	72
Na	65	32	0	0	97	91	61	32	93	77
K	6	1	0	0	7	8	1	1	2	6
Ca	12	1	0	0	13	15	3	1	4	11
Mg	16	7	0	0	23	24	14	7	21	14
Al	0	0	0	0	0	18	0	0	0	11
NH_4	57	0	6	0	63	16	0	6	6	6
NO_3	50	0	0	18	68	32	1	18	19	6
Cl	70	38	0	0	108	108	72	38	110	104
SO_4	86	4	8	13	111	115	8	25	33	58
Org.anion	0	0	0	0	0	18	0	0	0	28
Σ^+	220	41	8	31	300	274	89	80	169	197
Σ^-	206	42	8	31	287	273	81	81	162	196

Table 6b. Volume-weighted mean concentrations of major ions in wet precipitation (In) and runoff (Out) at EGIL and KIM catchments for the $1^1/_2$ year treatment period 13 June 1984 - 14 November 1985. Units: µeq/l.

	EGIL		KIM	
	In	Out	In	Out
H_2O	1346	1092	1102	835
H^+	48	93	9	86
(pH)	4.32	4.03	5.05	4.07
Na	48	83	55	92
K	4	7	1	7
Ca	9	14	3	13
Mg	12	22	13	17
Al	0	17	0	13
NH_4	42	14	0	7
NO_3	37	29	1	7
Cl	52	98	65	125
SO_4	64	105	7	70
Org.anion	0	16	0	34
Σ^+	163	251	81	236
Σ^-	153	250	73	235

About 75% of the incoming NH_4 and 50% of the incoming NO_3 are retained in the catchment (Table 6). NH_4 retention results from both biological uptake as well as ion exchange. NO_3 is retained by biological activity, but ion exchange is relatively unimportant for this mobile anion. The balanced SO_4 budget is an indication that the estimated dry deposit is approximately correct.

At KIM catchment the 1 1/2 year treatment has caused changes in the flux of H+, base cations, Al, NH_4, NO_3, and SO_4 (Table 6). Whereas EGIL catchment showed approximately equal flux of H+ out as in, reduction of H+ inputs at KIM have resulted in a net flux of H+ out of the catchment. Of the base cations both Na and Mg now show net accumulation in KIM catchment, perhaps due to a reversed "salt effect". The salt effect (Reuss and Johnson 1985) is the increase in base cation concentrations in soil solution as a result of an increase in anion concentration and cation exchange. With the lower concentrations of mobile anions SO_4 and NO_3 at KIM, concentrations of cations has also decreased. The budgets indicate that a portion of the incoming base cations is now retained in the catchment. The result may be a build up of the pool of exchangeable base cations on the soil. The net flux of Al is lower at KIM than at EGIL, perhaps because of the lower acid deposition and slightly higher pH levels in runoff at KIM due to treatment.

Treatment at KIM caused major decreases in NH_4 and NO_3 input, and output responded as well (Table 6). NH_4 deposition at KIM is only 10% of that at EGIL, and NH_4 outputs have declined such that output equals input. NO_3 inputs have also decreased, but not to such a great extent due to continued dry deposit of NO_2 gas. NO_3 outputs at KIM are now only 20% of those at EGIL. Whereas NH_4 and NO_3 were the third most important cation and anion, respectively, in both deposition and runoff at EGIL, at KIM they now are minor consituents.

The sulfate budget at KIM shows major differences from that of EGIL (Table 6). Whereas at EGIL SO_4 out equals SO_4 in, at KIM there is almost twice as much SO_4 leaving the catchment as enters in dry and wet deposition during the first 1 1/2 years of treatment. This period represents slightly less than one normal hydrologic year.

The SO_4 budget indicates a net loss of about 25 meq/m^2 from KIM. The measurements of Lotse and Otabong (1985) of water soluble SO_4, adsorbed SO_4 and SO4 in soil solution yield an estimate of about 40 meq/m^2 SO4 in the catchment. Thus about 2/3 of the readily-available SO4 has been lost from KIM as of November 1985.

DISCUSSION

At Sogndal response during snowmelt in 1985 was different than during snowmelt in 1984. pH levels in runoff from SOG2 declined to 4.1 in 1984 but only 5.0 in 1985 despite similar loading of acid to the snowpack. The difference may be related to snowpack volume or nature of the snowmelting.

Acid additions at Sogndal in 1984 caused increases in labile Al from natural levels of <50 µg Al/l to as much as 1000 µg Al/l (Figure 6). The data from 1984 indicated that an $Al(OH)_3$ solid phase with log ion activity product (3 pH-pAl^{+3}) of about 9.1 can account for labile Al concentrations at Sogndal (Figure 6) (Wright 1985). This solubility corresponds to that of synthetic gibbsite at 10^0C or natural gibbsite at 20^0C (Driscoll et al. 1984, Seip et al. 1984). Similar LAL-pH relationships are found elsewhere in Norway (Wright and Skogheim 1983, Seip et al. 1984) and in North America (Driscoll 1980, Driscoll et al. 1984, Johnson et al. 1981). The data from the second year of treatment (1985) indicate a significantly lower aluminum solubility.

Data from 1985 from both SOG2 and SOG4 suggest a log ion activity product of about 7.7. We postulate that the acid additions during 1984 at SOG2 and SOG4 resulted in the depletion of a reservoir of readily-soluble $AL(OH)_3$ with log solubility product of 9.1. Al precipitated in the stream channels may provide such a reservoir. We suggest that this Al, perhaps amorphous, occurs as a result of the natural increase in water pH as soil solution enters the stream and CO_2 degasses (Reuss and Johnson 1985). Our additions of strong acid in 1984 would then redissolve this $Al(OH)_3$ phase. By 1985 the reservoir is depleted, and Al concentrations in streamwater now reflect the Al solubility in soil solution. Here the Al comes from ion-exchange as well as weathering of primary and secondary minerals.

Labile aluminum levels at the highly acidified sites at Risdalsheia indicate log solubility product of about 6.8 (Wright 1985). It appears that Al solubility at the Sogndal catchments is moving towards the situation at Risdalsheia.

Sulfate concentration in runoff have responded differently to acid additions at Sogndal relative to acid exclusion at Risdalsheia. At the untreated control catchments at Sogndal the sulfate flux in equals the sulfate flux out. Concentrations in streamwater are about 25 µeq/l.

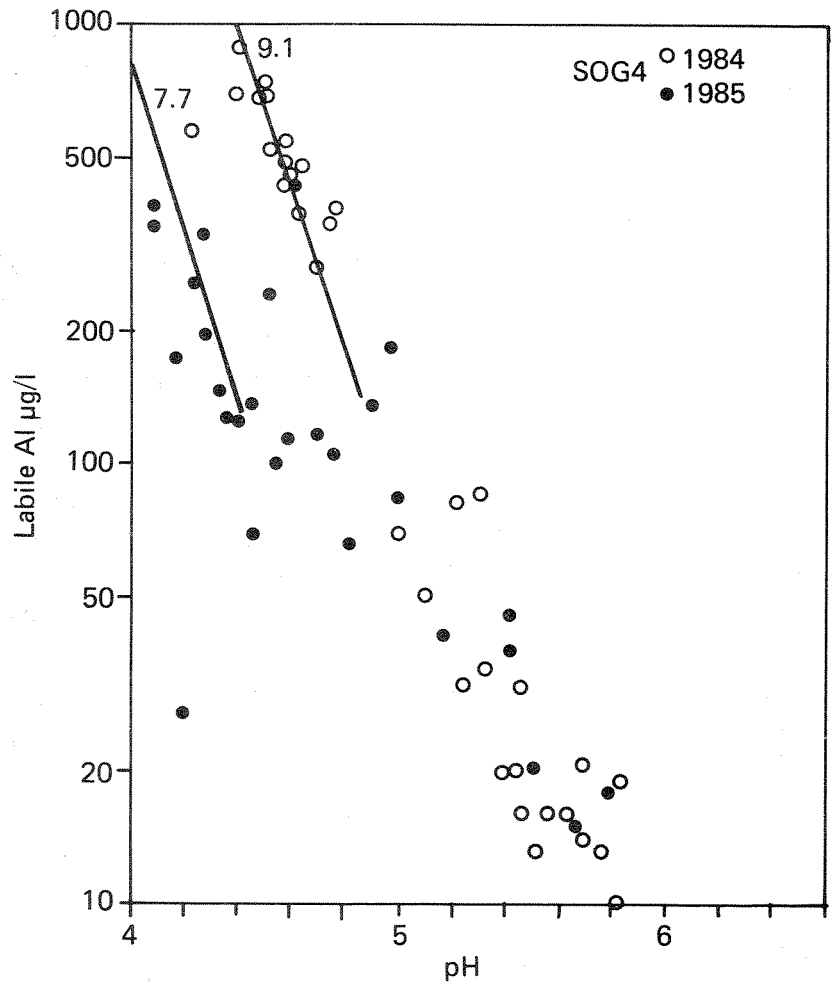
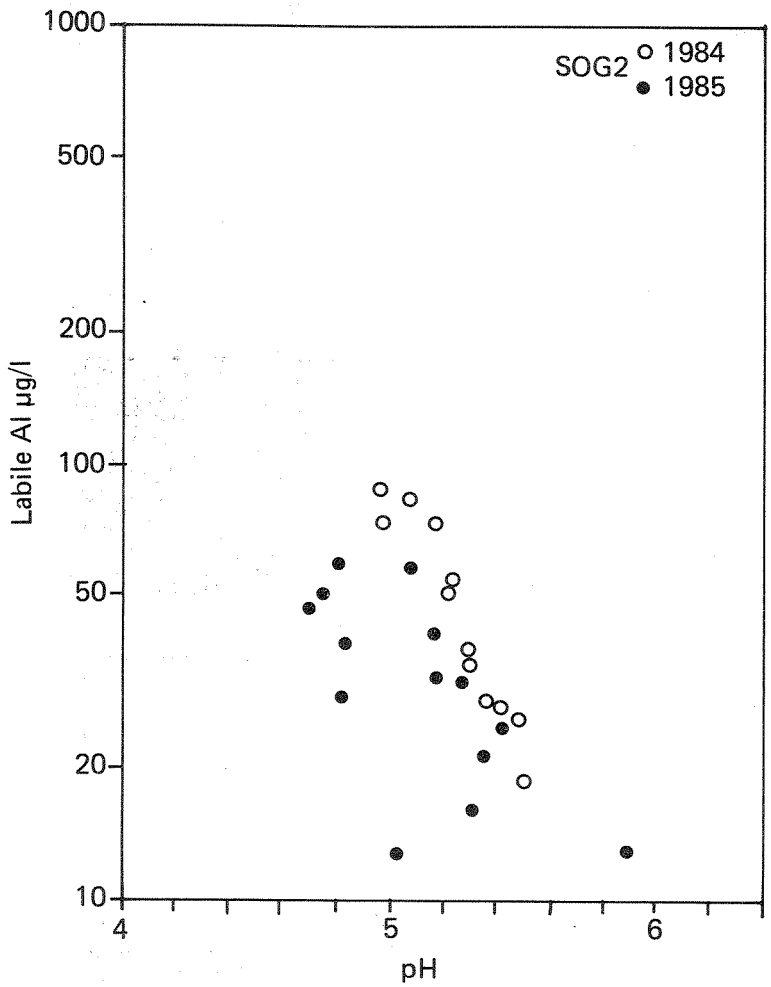


Figure 6a. Concentrations of labile aluminum and pH in runoff samples from Sogndal in 1984 (open circles) and 1985 (solid circles). Also shown are lines of equilibrium with $Al(OH)_3$ with log solubility product (3 pH-pAl^{+3}) of 7.7 and 9.1.

Figure 6b.



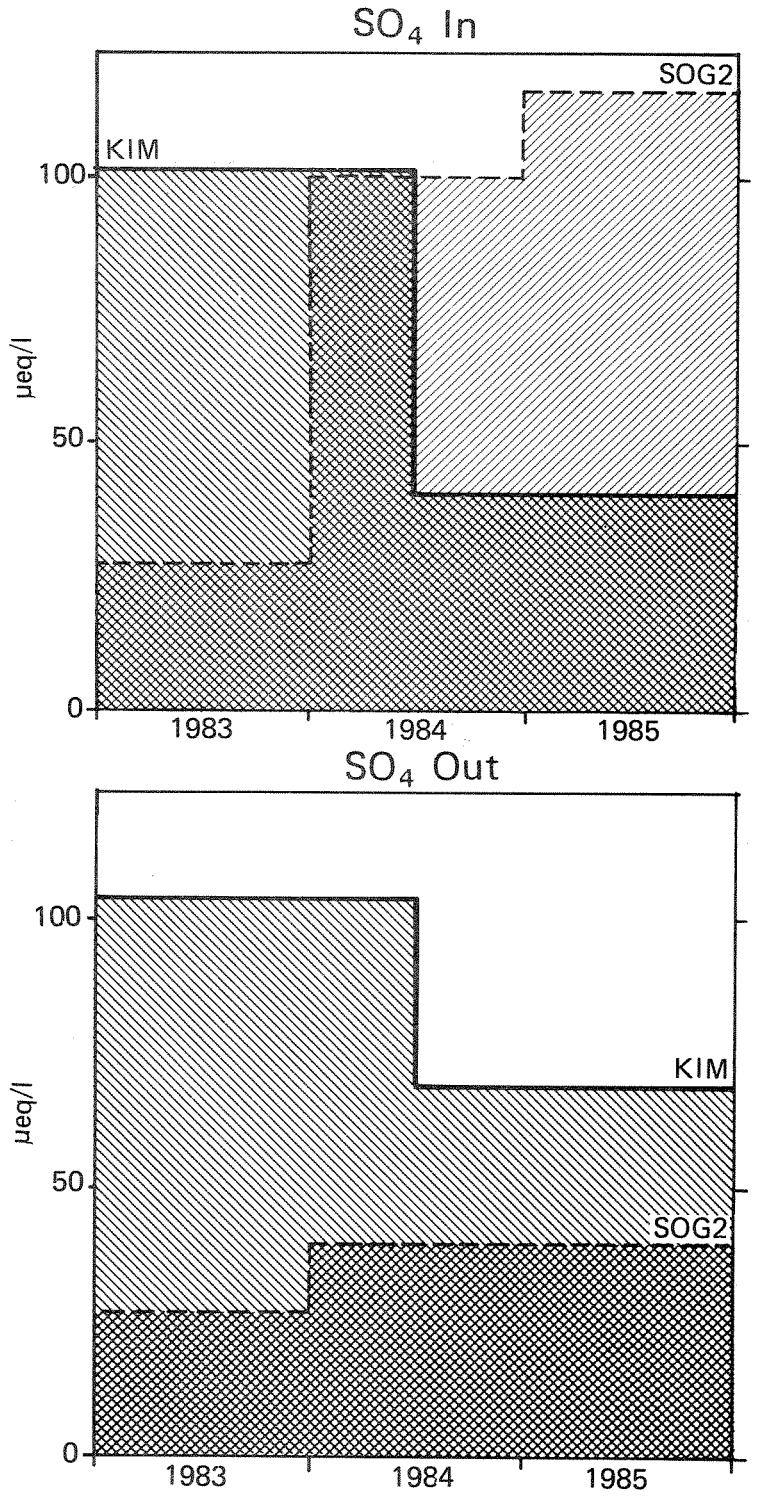


Figure 7. Volume weighted-average concentrations of sulfate in deposition and runoff at SOG2 (Sogndal) and KIM (Risdalsheia) before and after treatment. Concentrations in deposition are obtained by dividing input flux (wet and dry) by runoff volume thus adjusting for evapotranspiration.

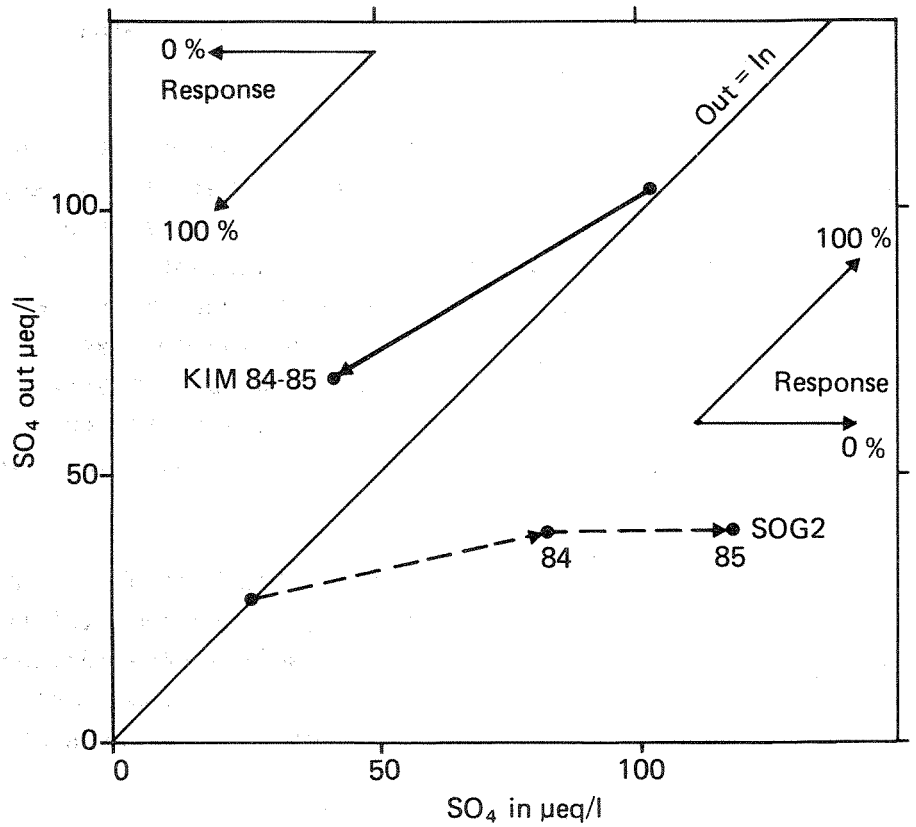


Figure 8. Response of sulfate concentrations in runoff to change in sulfate deposition at SOG2 (Sogndal) and KIM (Risdalsheia). Concentrations in deposition are obtained by dividing input flux (wet and dry) by runoff volume thus adjusting for evapotranspiration.

Acid addition to SOG2 increased the volume-weighted concentration in precipitation to about 100 µeq/l the first year and to about 120 µeq/l the second year. Runoff sulfate responded by increasing slightly to 40 µeq/l both years (Figure 7). Most of the sulfate is retained in the catchment, presumably by sulfate adsorption.

At Risdalsheia the control catchment EGIL also shows a balanced sulfate budget with inputs (wet and dry) equivalent to about 100 µeq/l in runoff and outputs about the same (Figure 7). The exclosure at KIM catchment resulted in reduction of inputs to the equivalent of about 40 µeq/l. Runoff response during the first 1 1/2 years has been a decline to about 70 µeq/l. The net loss of sulfate can be explained by the desorption of water-soluble sulfate in the soils, although other processes in the sulfur cycle such as mineralization and transformation from organic-S can also account for these data (Seip et al. 1979).

Response at Risdalshiea has thus been more rapid than that at Sogndal (Figure 8). This is consistent with the responses expected if the process of sulfate adsorption dominates in these soils. The response to an increase in sulfate deposition will be retention followed by a rapid "breakthrough", whereas the response to a decrease in deposition will be first rapid and then followed by a long "tail". This hysteresis is described by Reuss and Johnson (1986).

The RAIN project has provided valuable insight into the response of whole catchments to changes in loading of acids from the atmosphere. Although many of the results presented here are preliminary, we are hopeful that the RAIN project will help us understand the complex relationship between soil and water acidification and provide information useful for the establishment of target loadings and goals for future levels of emissions of acidifying compounds.

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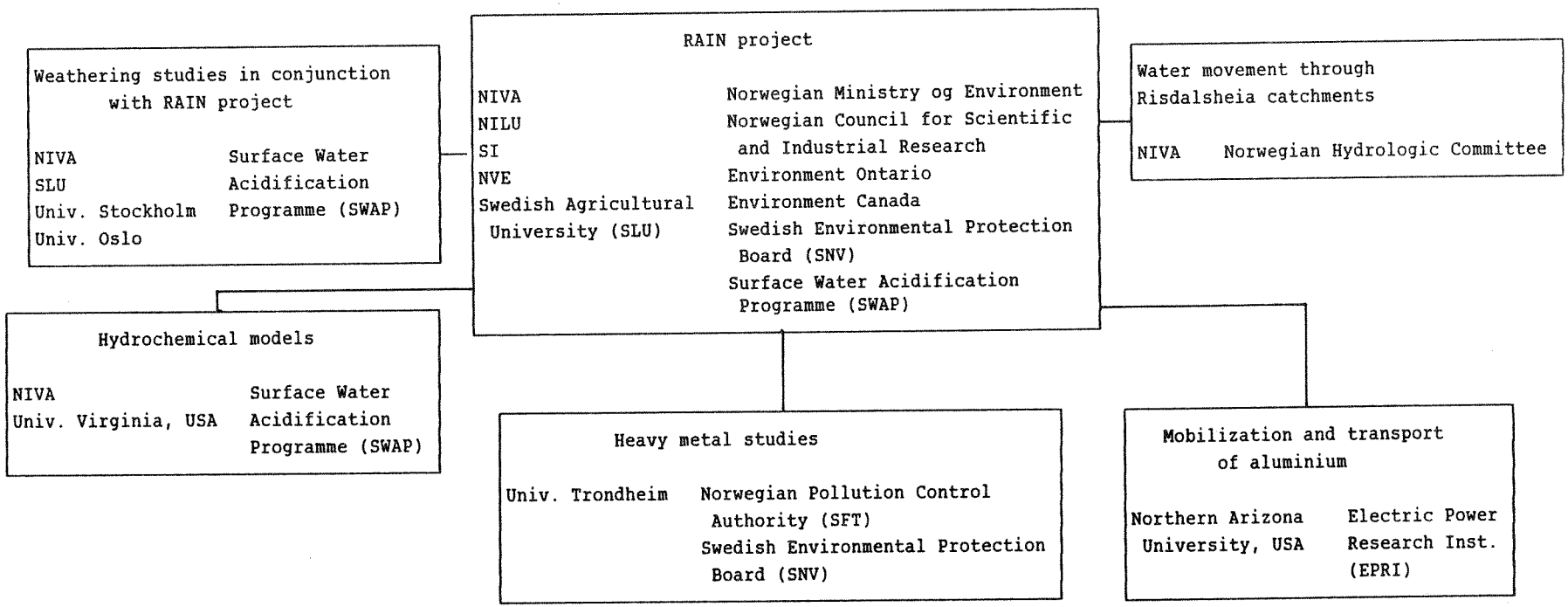
APPENDIX 1 VISITORS TO RISDALSHEIA IN 1985.

DATE	VISITORS AND AFFILIATION
2 MARCH	B.J. COSBY AND G.M. HORNBERGER, UNIVERSITY OF VIRGINIA, USA
19 APRIL	GROUP OF BIOLOGY STUDENTS, UNIVERSITY OF OSLO
23 APRIL	GROUP OF STUDENTS, GRIMSTAD GYMNAS
7 MAY	GROUP OF 50 4H MEMBERS FROM GRIMSTAD AND LILLESAND
28 MAY	F. LAST, N. CAPE, G.LEE, AND I. NICHOLSON, INSTITUTE OF TERRESTRIAL ECOLOGY, UK
30 MAY	ENGINEERING STAFF, AGDER INGENIOR DISTRIKTSHØYSKOLE
12 JUNE	GROUP OF 170 FOREST OWNERS (NORSK SKOGEIERLAG) ACCOMPANIED BY SCIENTISTS A. SEMB (NILU), G. ABRAHAMSEN (NISK), REPORTERS AND NORWEGIAN NATIONAL TELEVISION (NRK SØRLANDET)
20 JUNE	NIVAS BOARD OF DIRECTORS
21 JUNE	UNITED NATIONS WORLD COMMISSION FOR ENVIRONMENT AND DEVELOPMENT (BRUNDTLAND KOMMISSJON) ACCOMPANIED BY R. SURLIEN, MINISTER OF THE ENVIRONMENT, LOCAL POLITICIANS, AND REPORTERS
28 JUNE	R. PECHLANDER, UNIVERSITY OF INNSBRUCK, AUSTRIA
11 JULY	JOURNALIST FROM "NYTT FRA NORGE"
11 SEPT.	NORWEGIAN COUNTY FORESTERS
20 SEPT.	25 ENVIRONMENTAL OFFICIALS FROM AUST- AND VEST-AGDER COUNTIES
28 SEPT.	NATURE GROUP FROM BIRKELAND FOLKEHØYSKOLE
2 OCT.	14 ENGINEERING STUDENTS FROM AGDER INGERIOR DISTRIKTSHØYSKOLE
3 OCT.	H. HULTBERG, SWEDISH AIR AND WATER POLLUTION RESEARCH INSTITUTE, AND A JOURNALIST FROM "LAND"
24 OCT.	TELEVISION CREW FROM CANADIAN BROADCASTING CORPORATION

APPENDIX 2 RAIN PROJECT AND ASSOCIATED SATELLITE PROJECTS AS OF MAY 1986. Within each box are listed the participating institutions (left) and the funding agencies (right)



RAIN PROJECT

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Norwegian Institute for Water Research NIVA

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6/1984 Wright, R.F. 1984. Changes in the chemistry of Lake Hovvatn, Norway, following liming and reacidification. 68 pp.

7/1985 Wright, R.F. 1985. RAIN project. Annual report for 1984. 39 pp.

8/1985 *Lotse, E* and *Otabbong, E.* 1985. Physiochemical properties of soils at Risdalsheia and Sogndal: RAIN project. 48 pp.