

## **Effects of Acid Precipitation on a Small Acid Lake in Southern Norway**

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Langtjern is an acid lake situated in south-central Norway, 516 m above sea level. The watershed is underlain by biotite gneisses and granites. Coniferous forests cover 63% of the watershed, while 16% is covered by peaty areas. The lake has a relatively large watershed (4.8 km<sup>2</sup>) in relation to lake size (0.23 km<sup>2</sup>), and most of the water and chemicals reach the lake via the watershed and inflowing streams. Direct precipitation on the lake surface is less important.

The two major inflowing streams show rapid responses to influxes of acid precipitation (weighted annual mean pH 4.3) and long recovery periods between episodes. The fall and spring acid episodes have different impacts on the lake-water chemistry. The spring episode during which large amounts of H<sup>+</sup> are released in the first phases of the snow melt reduces the pH in only the surface layers of the still ice-covered lake and much of the inflowing pollutants are promptly discharged. In the fall, however, the acid inputs are readily mixed through the entire water column during the autumnal circulation period.

Assuming chloride as a conservative parameter, mass-balance calculations show that there is a net loss of Ca, Mg and Al from the watershed while there is a net uptake of H<sup>+</sup>, NO<sub>3</sub> and NH<sub>4</sub>. SO<sub>4</sub> and Na budgets balance. The net uptake of H<sup>+</sup> in the watershed is directly related to net losses of other cations. The net uptake of NO<sub>3</sub> and NH<sub>4</sub> is most likely due to biological uptake by the forest ecosystem.

For the Langtjern itself the inputs of all major ions equal the outputs, except for H<sup>+</sup> and NO<sub>3</sub> which are retained by the lake. The neutralization of H<sup>+</sup> in Langtjern varies seasonally, whereas only 10-20% is neutralized during spring, in the autumn 50-70% is neutralized.

## Introduction

A growing body of evidence suggests that the acidity of lakes and rivers in southern Scandinavia has increased over the past several decades. Chemical data collected in 1930-40 compared with data collected in the 1970's indicate that the pH in soft-water lakes has decreased over 1 pH unit; the acidity is increasing 5-25% per year (Wright and Gjessing 1976). Statistical analysis of pH-data in 7 major rivers in southernmost Norway (Sørlandet) indicate a doubling in acidity over the period 1965-75 (Henriksen et al., in prep.). In this part of Norway the majority of lakes and rivers are so acid that the natural fish populations are unable to survive (Jensen and Snekvik 1972, Leivestad et al. 1976). The cause of the acidification of these freshwaters is most probably the deposition of acid precipitation which primarily originates from the emission, oxidation and long-range transport of sulfur and nitrogen oxides released from the burning of fossil fuels (Dovland et al. 1976).

Norwegian freshwaters are particularly vulnerable to acid precipitation because much of southern Norway is characterized by granitic bedrock only partially covered by thin glacial deposits of similar mineralogy and podzolic soils. Chemical weathering proceeds slowly, and thus surface waters typically have low concentrations of major ions and very low buffer capacities (Gjessing et al. 1976).

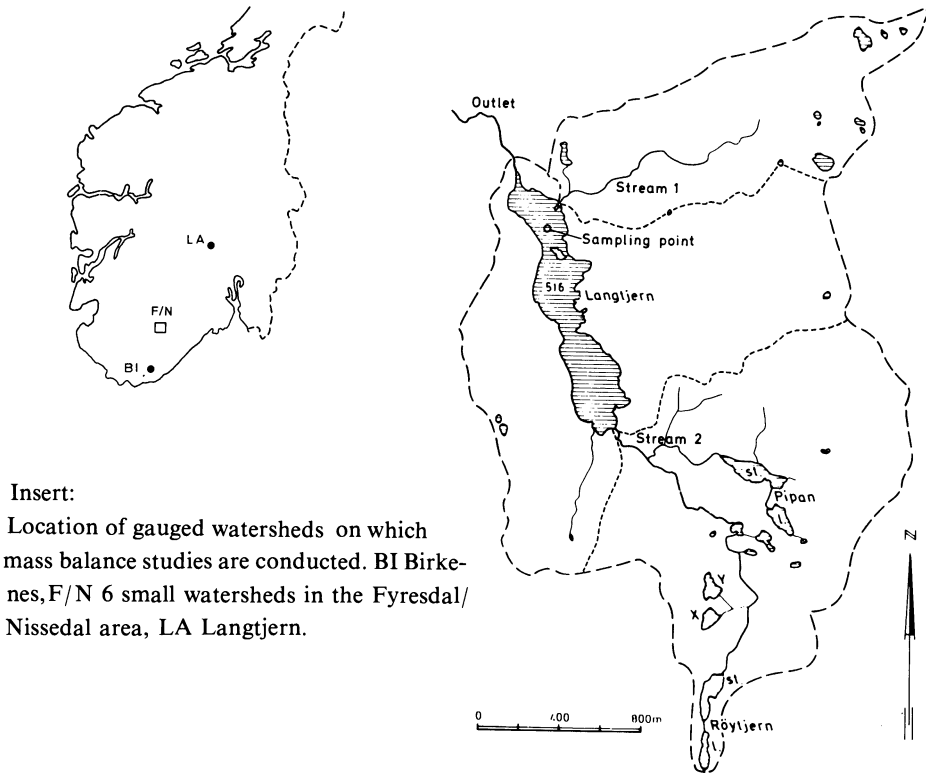
Because of the increasing concern about the adverse effects of acid precipitation in Norway, a major research project entitled »Acid Precipitation - Effects on Forest and Fish« (the SNSF-project) was launched in 1972. As part of the SNSF-project intensive research on hydrology and hydrochemistry is underway at 10 gauged watersheds and lake basins in southern Norway. We present here results from studies at one of these watersheds, a small acid lake called Langtjern (LA), situated in south-central Norway about 100 km NW of Oslo (Fig. 1).

## Description of Langtjern and its Watershed

Langtjern (Fig. 1) is an acid dystrophic lake, 0.23 km<sup>2</sup> in area, maximum depth of 8 m in the north basin, mean depth of 2 m, volume of 0.56 x 10<sup>6</sup> m<sup>3</sup>, and a theoretical water-retention time of 58 days (volume/mean outlet discharge). Langtjern's water chemistry (Table 1) is typical of small acid lakes in southern Norway (Gjessing et al. 1976, Wright and Henriksen, in prep.). Langtjern lost its natural fish population in the early 1960's, probably because of acidification due to acid precipitation.

The watershed of Langtjern (4.8 km<sup>2</sup>) is underlain by biotite felsic gneisses and granites with patches of ground moraine of generally the same lithology. Coniferous forests of *Pinus sylvestris* and *Picea abies* cover 63% of the watershed, 16% is covered by peaty areas, 5% is water and 16% is bare rocks.

Two permanent streams drain well-defined portions of the watershed (Fig. 1). These streams deliver about 60% of the runoff to Langtjern, while the remainder comes via surface or interflow in ephemeral streams and rivulets.



Insert:  
Location of gauged watersheds on which mass balance studies are conducted. BI Birkenes, F/N 6 small watersheds in the Fyresdal/Nissedal area, LA Langtjern.

Fig. 1. Map of Langtjern and its watershed showing the two major inflowing streams, the outlet and the sampling point in the lake.

## Methods

Hydrological and chemical studies at Langtjern were started in May 1973 when the outlet stream was equipped with a V-notch weir and instrumentation for continuous discharge measurements. Water samples for chemical analysis are collected weekly from the two inflowing streams and the outlet stream (Fig. 1). Samples are taken manually with 1/2-l polythene bottles, previously washed with Na-EDTA-solution, nitric acid and distilled water.

The sampling of Langtjern itself started in January 1974. Samples are collected from depths of 1, 2, 4, and 6 meters in the north basin. The sampling frequency varies according to expected changes in water quality. During the winter stagnation period the lake is sampled once a month. During the summer stagnation period the frequency was once a week in 1974 and once every second week in 1975. During the snow melt in spring and the fall storms the sampling frequency is increased to several times a week.

Table 1 - Chemical data, Langtjern. Arithmetic mean and standard deviation for samples collected at 1-m depth over the period 23 January 1974 to 10 October 1976.

Components		No. of observations	Mean	Standard deviation
pH		90	4.95	0.23
Conductivity	$\mu\text{S}/\text{cm } 20^\circ\text{C}$	90	17.7	3.1
Color	mg Pt/l	86	100	30
Ca	mg/l	88	1.38	0.27
Mg	"	89	0.76	0.04
Na	"	74	0.70	0.18
K	"	89	0.17	0.06
SO <sub>4</sub>	"	84	3.8	0.8
Cl	"	86	0.68	0.16
NO <sub>3</sub> -N	$\mu\text{g}/\text{l}$	89	24	25
NH <sub>4</sub> -N	"	19	25	25
Total-P	"	11	7	3
Al	"	73	218	46
Fe	"	9	184	44
Zn	"	15	15	7
Pb	"	11	2.0	0.8
Cu	"	7	6	4
Cd	"	3	0.21	0.10

Precipitation is sampled daily for chemical analysis at Gulsvik, 7 km from Langtjern. For comparison weekly precipitation samples are collected at Langtjern during the summer months. Precipitation, temperature and humidity are continuously recorded at the site.

Chemical analysis of the samples proceeds according to the routine methods used at the Norwegian Institute for Water Research (Wright and Henriksen, in prep.). The analytical program is shown in Table 2.

Errors in the raw data arising from labelling and transcription mistakes and mistakes in punching computer cards are uncovered and corrected by 1) calculating the ion balance of each sample and 2) calculating the theoretical electrical conductivity from the contribution of each ion and comparing the theoretical with the measured conductivity of each sample.

## Results and Discussion

### Precipitation

Langtjern is situated in the region of southern Norway that receives highly acidic precipitation (Dovland et al. 1976). Wet precipitation at Langtjern has a weighted-mean pH of 4.32 (July 1974 - June 1975), similar to values of pH 4.34 and 4.29 measured at Nissedal/ Fyresdal and Birkenes, two other SNSF-project watersheds

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Table 2 - Analytical program for stream, lake and precipitation samples from Langtjern.

Component	Streams	Lake	Precipitation
pH	x	x	x
Conductivity	x	x	
Color		x	
Nitrate	x	x	x
Ammonia			x
Orthophosphate			x
Sulfate	x	x	x
Chloride	x	x	
Potassium	x	x	x
Sodium	x	x	
Calcium	x	x	x
Magnesium	x	x	x
Aluminium	x	x	
Zinc			x
Temperature		x	

located in southernmost Norway (Fig. 1) (Gjessing et al. 1976). (The weighted-mean pH is calculated by multiplying the volume of precipitation by the hydrogen-ion concentration in each sample as derived from pH measurements, adding these daily H<sup>+</sup>-depositions over the entire year and then dividing by the total precipitation volume for the year). At Langtjern, as is generally the case in southern Norway, precipitation contains approximately equal amounts (on an equivalent basis) of sulfate and hydrogen ions and nitrate and ammonia ions (Dovland et al. 1976). These four components dominate the chemistry of precipitation. Minor amounts of the major cations Ca, Mg, Na, and K and of Cl are also present in precipitation, generally in the same relative concentrations as in seawater, indicating that seawater spray is the major source of these ions.

### Lake and Stream Water Chemistry

At Langtjern the chemical composition of the inflowing streams and the lake itself varies in response to episodic inputs of acid precipitation. As an example the pH observations in the two inlet streams and in the lake itself are presented in Fig. 2 together with the outlet discharge and the H<sup>+</sup> deposition through precipitation. The two inflowing streams exhibit a pattern of rapid response to acid-precipitation episodes with long recovery periods between episodes. The fall and spring episodes have different impacts on the lake-water chemistry. The spring episode, during which massive amounts of H<sup>+</sup> are released during the first phases of snowmelt (Henriksen et al. 1974, Johannessen et al. 1975), reduces the pH only in the surface layers of the still ice-covered lake. The inflowing meltwater is less dense than the 4°C bottom waters and

moves across and out of the lake in the surface water. Thus much of the incoming pollutant load is promptly discharged. In the autumn, however, the acid inputs come during the autumnal circulation period and are readily mixed through the entire water column. As with the stream water, recovery to pre-episode levels also proceeds slowly in the lake itself. These rapid drops in pH during spring and autumn are particularly critical for fish, because this is when spawning and hatching take place, and it is the reproductive process that appears most sensitive to acid stress (Leivestad et al. 1976).

### **Mass-Balance Calculations**

Mass-balance data for Langtjern and its watershed for the periods July 1973 - June 1974 and July 1974 - June 1975 are displayed in Table 3. The chloride data show that in 1973-74 1.4 and in 1974-75 1.5 times more Cl left the watershed in the stream water than was measured in the incoming precipitation (wet-only). Measured wet-precipitation underestimates total deposition because precipitation gauges generally do not collect 100% of the precipitation volume, and because dry-deposition is not measured. Since neither the rocks nor the soils in the Langtjern watershed provide a significant primary source of Cl, over the long-term Cl outputs should approximately equal inputs. Thus based on the reasoning given by Gjessing et al. (1976), a Cl balance was assumed and the inputs of all other components were scaled-up by the same factor as for Cl (Table 3).

The mass balance data for the lake and watershed show that (1) inputs approximately equal outputs for Cl (assumed), Na,  $\text{SO}_4$  and K, (2) there is a net uptake of  $\text{H}^+$ ,  $\text{NO}_3$  and  $\text{NH}_4$ , and (3) there is a net loss of Ca, Mg and Al. The retention of  $\text{NO}_3$  and  $\text{NH}_4$  is most probably due to biological uptake of these nutrient compounds.

The loss of Ca, Mg and Al from the watershed may partly be due to natural weathering processes, but the major loss is probably due to the inputs of acid precipitation. The retention of  $\text{H}^+$  apparently produces losses of other cations, possibly through cation exchange reactions in the soil. Likens and Borman (1975) reached a similar conclusion based on mass-balance data from undisturbed forested watersheds at Hubbard Brook Experimental Forest, New Hampshire, USA, an area that also receives highly acidic precipitation (pH 4.1).

The principal features of the chemical budget data for Langtjern and its watershed also characteristic for 6 small watersheds in the Fyresdal/Nissedal area and for the Birkenes watershed (Fig. 1) (Gjessing et al. 1976).

The calculated ionic balance for the chemical budget is also shown in Table 3. Aluminium is not included in the ionic balance, because this element is probably not present as a free ion at the pH 4.8-5.0 levels in Langtjern. The difference between cations and anions is positive for both precipitation and the outlet waters and the percent difference is similar for both years. This difference may be due to either an overestimate of cations or an underestimate of anions. The contribution of the bicarbonate ion (not measured) has been calculated to account for less than 1 kiloequivalent/ $\text{km}^2/\text{yr}$ . and cannot explain the difference. Since the concentrations of Ca, Mg,

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Table 3 - Chemical budgets for Langtjern and its watershed. Unit: kiloequivalents/km<sup>2</sup>/yr.

	1 July 1973 - 30 June 1974						1 July 1974 - 30 June 1975					
	Wet precipitation only			Total inputs <sup>a</sup>			Wet precipitation only			Total inputs <sup>a</sup>		
	In	Out	Out/In	In	Out	Out/In	In	Out	Out/In	In	Out	Out/In
Water (mm)	520	380	0.73				800	735	0.92			
H <sup>+</sup>	29	7.5	0.26	41	7.5	0.18	38	16	0.42	58	16	0.27
Na	5.5 <sup>b</sup>	8.3	1.5	7.8	8.3	1.1	11 <sup>b</sup>	17	1.5	16	17	1.1
K	1.4	1.7	1.2	2.0	1.7	0.8	2.0	2.3	1.2	3.0	2.3	0.8
Ca	4.5	26	5.8	6.2	26	4.2	6.4	43	6.7	9.8	43	4.4
Mg	1.5	8.3	5.6	2.2	8.3	3.8	2.9	14	4.8	4.5	14	3.0
Al <sup>c</sup>	0.6 <sup>d</sup>	8.3	14	0.8 <sup>d</sup>	8.3	10	0.9 <sup>d</sup>	16	18	1.4 <sup>d</sup>	16	11
SO <sub>4</sub>	32	34	1.1	45	34	0.8	41	59	1.4	63	59	0.9
Cl	4.7	6.7	1.4	6.7	6.7	1.0	9.2	14	1.5	14	14	1.0
NO <sub>3</sub>	15	0.9	0.06	22	0.9	0.04	21	1.2	0.06	32	1.2	0.04
NH <sub>4</sub>	14	0.9 <sup>e</sup>	0.06	19	0.9 <sup>e</sup>	0.05	17	1.2 <sup>e</sup>	0.07	26	1.2 <sup>e</sup>	0.05
∑ cations	H <sup>+</sup> , NH <sub>4</sub> <sup>+</sup> , Ca <sup>2+</sup> , Mg <sup>2+</sup> , K <sup>+</sup> , Na <sup>+</sup>			79	53					117	93	
∑ anions	(SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> )			74	42					109	74	
»Excess« cations				5	11					8	19	
»Excess« in percent				7	21					7	20	

a. Dry deposition estimated assuming Cl in = Cl out.

b. Calculated from Mg inputs and Na/Mg ratio in seawater.

c. Calculated assuming all Al present as Al<sup>3+</sup>.

d. Estimated from representative samples of precipitation.

e. Calculated assuming [NH<sub>4</sub>-N] = [NO<sub>3</sub>-N].

Na, and K are determined by atomic-absorption spectrophotometry, a method that gives the total amount present, a fraction of the measured amounts may be in the form of electrically-neutral complexes, colloids, or particles and thus do not contribute to the ionic balance. In precipitation fractions of Ca and Mg may be present in particulate matter, and in the lake water there is strong evidence that fractions of the Ca and Mg may be bound by humic compounds (Gjessing 1976).

pH-data from the outlet of Langtjern and from the inflowing streams indicate that a substantial part of the acid entering the lake is neutralized in the lake itself. Although the inflowing streams are not gauged, the flow in these streams can be estimated using a simple model and the outlet discharge data. These estimated flow data can then be combined with the chemical concentration data, and approximate chemical budgets for Langtjern itself are obtained (Table 4).

These budgets indicate that only H<sup>+</sup> and NO<sub>3</sub> are significantly affected in the lake. All other chemical components apparently leave the lake in the same amounts as enter the lake. Biological processes in the lake such as denitrification or nitrate uptake probably account for the net loss of nitrate in Langtjern; an equivalent amount of H<sup>+</sup> will be lost by these processes (Brewer and Goldman 1976).

Table 4 - Chemical budget for Langtjern during the period 1 July 1974 - 30 June 1975. Unit: kiloequivalents/km<sup>2</sup>/yr.

	In		Total	Out	Out/In
	Runoff from watershed	Precipitation on lake			
H <sup>+</sup>	22	2.9	25	16	0.64
Na	18	0.8	19	17	0.90
K	2.2	0.1	2.3	2.3	1.0
Ca	40	0.5	40	43	1.1
Mg	14	0.2	14	14	1.0
Al	16	-	16	16	1.0
SO <sub>4</sub>	61	3.1	64	59	0.92
Cl	14	0.7	15	14	0.94
NO <sub>3</sub>	0.8	1.6	2.4	1.2	0.50
			In	Out	Loss
∑ cations (Al excluded)			100	92	8
∑ anions			81	74	7
»Excess« cations			19	18	1

The neutralization of incoming acid in Langtjern varies seasonally. The data in Fig. 2 show that the lake responds differently to the spring and autumn inputs of acid. Splitting the annual H<sup>+</sup>-budget into two parts, January-June and July-December, provides an estimate of the amounts of acid neutralized in the lake during the spring and fall episodes, respectively (Table 5).

Whereas only 10-20% of the acid entering the lake in the spring is neutralized, in the autumn 50-70% is neutralized, probably because the autumn inputs come during lake circulation and are mixed with the bottom waters. The inputs during snowmelt in the spring, however, apparently do not mix with the bottom waters and merely pass out of the lake in the surface waters (Fig. 2).

Table 5 - H<sup>+</sup>-balance for Langtjern, spring and fall episodes. Unit: kiloequivalents/km<sup>2</sup>.

	1974		1975	
	Jan.-June	July-Dec.	Jan.-June	July-Dec.
In (Total)	8.0	12.7	10.3	5.1
Out	6.5	6.4	9.0	1.5
Loss in lake	1.5	6.3	1.3	3.6
Loss in percent	19	50	13	71



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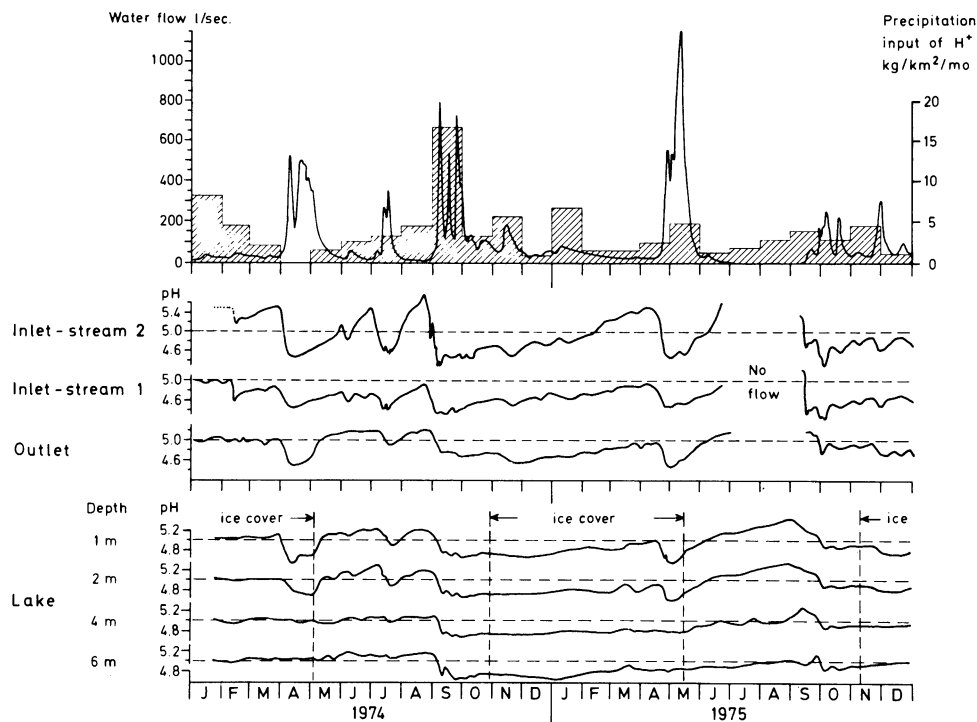


Fig. 2. The annual pH variations in the 2 inflowing streams, outlet and north basin of Langtjern in relation to outlet discharge and monthly deposition of H<sup>+</sup> in wet-precipitation.

Chemical data for Langtjern and its watershed and for Langtjern itself indicate that the long-term deposition of acid precipitation has apparently overwhelmed the natural buffer capacity of both the watershed and the lake.

The terrestrial watershed, characterized by bedrock and soils highly resistant to chemical weathering, neutralizes 75% of the incoming acid, but because precipitation has a weighted-average pH of 4.3, the two streams are still highly acid, weighted-average pH 4.6. These streams in turn deliver acid to Langtjern itself, where only 40% is neutralized in the lake; the outlet has weighted-average pH 4.8. Because the acid inputs are episodic, the acidity of both streams and the lake varies dramatically during the year. The periods of highest acid inputs, spring and autumn, are thus the periods of highest acidity in the streams and lake, and coincidentally the periods most critical for fish and other aquatic organisms. That Langtjern lost its natural fish population in the early 1960's attests to the acidification of the lake. Fish populations have been affected in similar poorly-buffered lakes in a 20,000 km<sup>2</sup> area of southern Norway (Leivestad et al. 1976). Chronic inputs of acid precipitation are probably responsible for the acidification of these freshwaters and subsequent loss of fish populations.

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