

## Research Article

# Increased colour and organic acid concentrations in Norwegian forest lakes and drinking water – a result of increased precipitation?

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**Abstract.** Recently there have been major increases in water colour and dissolved organic carbon (DOC) in many forest lakes that serve as drinking water reservoirs in the southern part of Norway. We studied changes in the physical and chemical properties in (I) a selected group of lakes and (II) raw water from the municipal waterworks of Oslo since 1983 and 1976, respectively. The largest increase in DOC and water colour occurred between 1997 and 2001. Statistically significant correlations are seen between water colour and accumulated amounts of precipitation, while variation in temperature had no apparent

effect. Despite significant reduction in sulphate concentrations in precipitation and lake waters since 1980, the lakes were more acidic in 2000 and 2001 than in previous years. Increases in colour and acidity, together with somewhat minor changes in DOC concentrations, indicate changes in the physical and chemical properties of the natural organic matter (NOM). Alteration in properties of NOM is most probably caused by increased precipitation that has changed the water pathways in the catchments and increased the leaching of organic components from the upper forest floor.

**Key words.** Climate change; water colour; NOM; organic acids; sulphate; acidification

## Introduction

Climate variations have caused unusual weather conditions in Northern Europe during the latest years. In Scandinavian countries, increased temperature and precipitation have been reported for the last century (SMHI, 2002; Hyvärinen, 2003), where especially the late 1980s and 1990s have received more precipitation than normal (SMHI, 2002; Hole and Tørseth, 2002). Scenarios developed by REGCLIM (Regional Climate Development Under Global Warming) for future climate in Norway predict both long-term increases in temperature and changes in

precipitation, as well as increased frequency and severity of extreme events such as floods and storms. In periods with high precipitation, alteration in hydrological flow patterns might occur. Enduring rain causes saturated soils with increasing flow through upper organic soil horizons (e.g. Steinberg, 2003). This effect causes changes in the fluxes and chemical composition of dissolved natural organic matter (NOM) compared to low flow conditions (Riise and Salbu, 1994; Hinton et al., 1997; 1998; Schiff et al., 1998; Riise, 1999; Hongve, 1999). Changes in the quantity and quality of dissolved NOM will subsequently influence physical and chemical parameters in surface waters (Ivarsson and Jansson, 1994; Scott et al., 1998; Laudon et al., 1999; Correll et al., 2001). Of major concern is the acidification status of lakes because NOM contributes to the acidity of surface waters (Brakke

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et al., 1987). A change in climate might, therefore, imply large scale impacts on the chemical composition of surface waters.

Anthropogenic acidification of aquatic ecosystems, caused by long-range transport of atmospheric pollutants, has been one of the main environmental issues in Scandinavia since the 1970s. Major national and international efforts have been made to reduce emissions. During the period 1980–2001 the content of sulphate in precipitation in Norway decreased by 63–72% (Aas et al., 2001). However, to evaluate the effects of reduced atmospheric deposition on lake water chemistry, better knowledge of the simultaneous response to increased precipitation and changing hydrological flow patterns is needed.

The water colour ( $\text{mg l}^{-1}$  Pt) increased in Swedish lakes and rivers between 1965 and 1986 (Forsberg and Petersen, 1990; Andersson et al., 1991). The colour was variable or even decreasing up to the middle of the 1970s and the increase was strongest towards the end of the period (Andersson et al., 1991). Increased precipitation in the 1980s (Karlström, 1999) was responsible for the greatest changes, but other mechanisms of an increasing greenhouse impact also were discussed (Forsberg, 1992). More recently, reduced input of NOM from terrestrial catchments has been ascribed to climatic warming and drought (Schindler et al., 1997), while global warming was the supposed reason for increased concentrations of DOC in runoff from upland catchments in the UK (Freeman et al., 2001).

Regarding the quality of NOM, it was shown that the ratio between colour and DOC in surface waters from all over southern Norway increased with increasing hydraulic discharge (Hongve and Andersen, 1991).

Norwegian waterworks and health authorities have recently noticed a significant increase in water colour in many lakes used for drinking water purposes. Some major waterworks that serve the capital area have experienced more than a doubling in raw water colour during 1997–2001 (Liltved et al., 2001). Increasing water colour is of hygienic and economic importance since it interferes with common disinfection procedures and necessitates more comprehensive water treatment to comply with drinking water standards.

In this paper we present results from lakes in a forest area bordering the city of Oslo. A selected group of pristine lakes have been studied on various occasions since 1983 (Riise and Hongve, 1987; Riise et al., 2000a). Water samples collected from the surface layer in late autumn is regarded to provide a good picture of the acidification status (Henriksen et al., 1988). In addition, long-term monitoring data from one of the larger lakes in this area is available, as the lake is used as raw water reservoir for the municipal waterworks of Oslo. Some lakes in the first group are within the catchment of this reservoir while others belong to neighbouring or more remote catch-

ments. However, we assume that large scale environmental impacts are relatively constant for all the lakes. Our aim has been to analyse the two separate sets of data to see if changes in water chemistry, acidification status and organic impact are interrelated, and to what extent they can be explained by climatic variables.

## Material and methods

The lakes in this study are situated in Østmarka, a forest area bordering the Oslo city district ( $59^{\circ}\text{N}$ ,  $10^{\circ}\text{E}$ ) in a southeast direction (Fig. 1). The area is 5–25 km from the city centre and is used for recreation and forestry. The elevation within the area varies from 125 to 395 m above sea level. At a nearby meteorological station (Oslo-Blindern), the mean precipitation (1961–1990) is  $763 \text{ mm y}^{-1}$ , ranging from 531 to  $1173 \text{ mm y}^{-1}$  during the study period. Annual mean temperature is  $5.7^{\circ}\text{C}$ . Figure 2 shows annual variations as differences from the 1961–1990 mean values (Statistical yearbook of Norway, 1976–2002). The lakes are on a bedrock of granite and gneiss with a thin cover of glacial deposits. In areas with sufficient soil thickness, podzols are predominant together with mires. The surface waters in the area are generally dilute (conductivity  $2.5\text{--}4.0 \text{ mS m}^{-1}$ ) with a marked influence of coloured NOM from terrestrial sources. The first survey from 1983 included 56 localities selected to be representative of all major lakes and lake complexes in the area. Since many of these lakes subsequently have been subject to liming, the 24 lakes used for the present study were carefully selected to include only pristine and non-limed localities. The mean lake surface area is  $0.2 \text{ km}^2$  (range  $0.005\text{--}0.8 \text{ km}^2$ ) and catchment area  $2.5 \text{ km}^2$  ( $0.1\text{--}9 \text{ km}^2$ ). Water samples were collected from the surface layer (0–2 m) or lake outlets during the autumn circulation period in 1983, 1996, 1997, 2000 and 2001. Chemical analy-

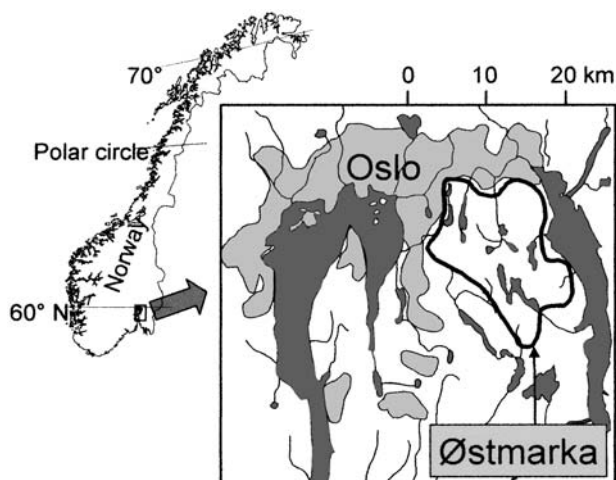
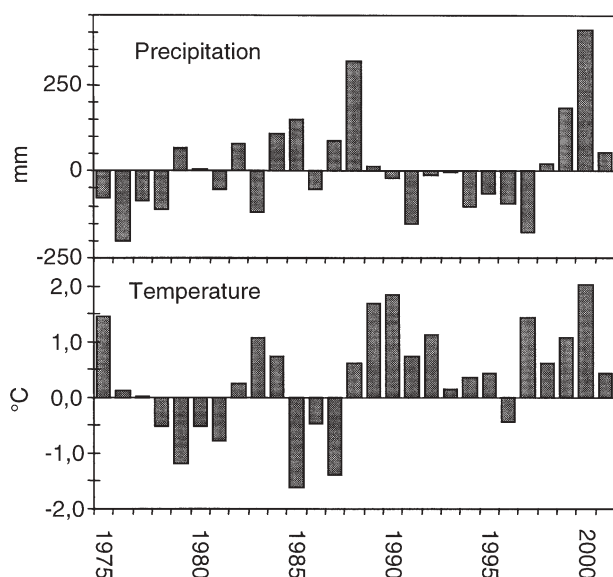


Figure 1. Location of the Østmarka area (encircled) outside Oslo.



**Figure 2.** Residuals for annual precipitation and mean temperature relative to the average values for precipitation (763 mm  $y^{-1}$ ) and temperature (5.7°C) for Oslo, 1961–1990.

ses were made in 1983 by Oslo Water and Sewage Works, in 1996 by the Agricultural University of Norway, Dept. of Soil and Water Sciences and in 1997–2001 by the accredited water laboratory of the Norwegian Institute of Public Health. In 1983 DOC was determined with a Xertex Dohrman DC 80 Organic Carbon Analyser and in 1997–2001 with a Scalar San plus segmented flow analyser. Both instruments are based on oxidation with persulfate and UV radiation and measurement of the formed  $CO_2$ . In 1996 these analyses were made with a Shimadzu TOC 5000 carbon analyser based on high temperature combustion. Results of the latter analyses have been adjusted for high blank values (0.75 mg  $l^{-1}$ ). As all samples were collected in oligotrophic lakes in a season with low productivity, the contribution from autochthonous C sources are regarded small. Furthermore, as studies from the investigated lakes have shown that the concentration of organic carbon in nonfiltered and membrane filtered samples (pore size 0.45  $\mu m$ ) were not significantly different ( $p < 0.01$ ) and that the sample injector on the carbon analyzer discriminates against particles, the samples have not been subject to filtration. The results on organic carbon have, therefore, been denoted as dissolved organic carbon (DOC). Water colour was measured spectrophotometrically according to the national standard method (The Norwegian Standards Association, 2002; Hongve and Åkesson, 1996). Alkalinity (in the text often denoted as  $HCO_3^-$ ) has been determined by titration with a strong acid (HCl) to pH 4.5 (The Norwegian Standard Association 1981a). In addition, the acid neutralizing capacity (ANC) has been determined as the difference between the sum of the basic cation concentrations ( $C_B = [Ca^{2+}] +$

$[Mg^{2+}] + [Na^+] + [K^+]$ ) and strong acid anion concentrations ( $C_A = [NO_3^-] + [SO_4^{2-}] + [Cl^-]$ ) ( $ANC = C_B - C_A$ ).

Oslo Water and Sewage Works have analysed raw water samples from Skullerud water treatment plant. The water is extracted from 40 m depth in one of the larger lakes in Østmarka, Lake Elvåga with a surface area of one  $km^2$  and theoretical water retention time of around two years. Lake Elvåga was not included in the investigated group of 24 lakes. Water colour and chemical oxygen demand ( $COD_{Mn}$ ) have been analysed since 1976. However, the quality of the data for  $COD_{Mn}$  was significantly improved in 1982 when a new national standard became available (The Norwegian Standards Association, 1981b). We have, therefore, excluded all previous data for this parameter. Data on organic carbon are available from March 1986, but results obtained before September 1988 show monthly fluctuations far beyond what is realistic for the water source. We assume this is due to analytical problems and have decided not to consider these results. An Astro 1850 TOC analyser based on UV-persulfate oxidation was in use from 1986, while newer instruments, Astro 2100 since 1996, and Shimadzu 5000A since 1999, are based on high-temperature combustion.

A t-test (calculation performed by SIGMAPLOT Jandel Scientific software) was used to calculate whether differences were statistically significant ( $p < 0.01$ ). Linear regression analyses were performed by Statgraphics version 5.0.

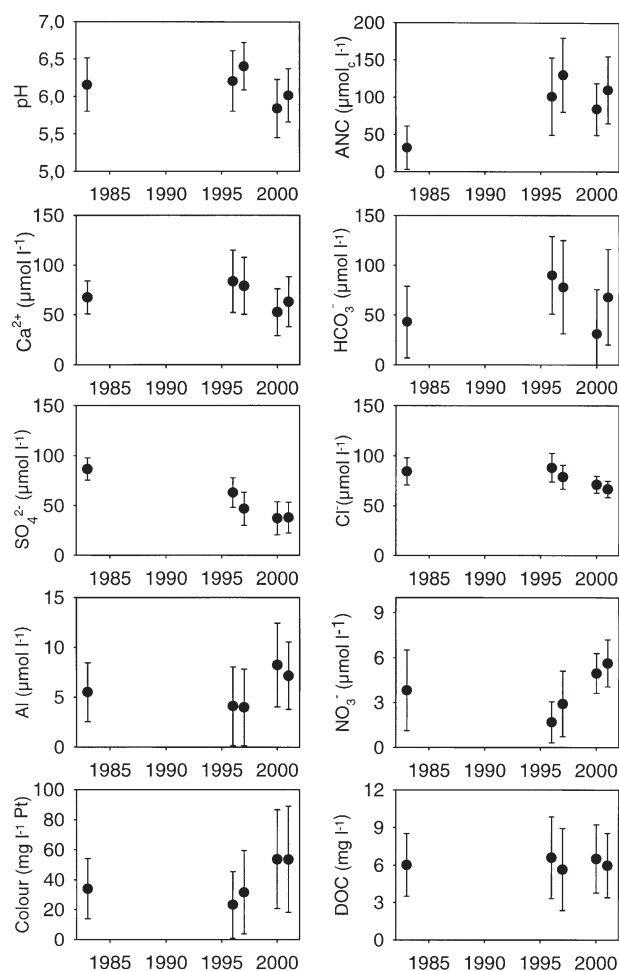
## Results

A comparison of physical and chemical data from the 24 selected lakes with the original assembly of 56 localities shows that the selection is still representative for the area except for the most acidic lakes that were excluded due to liming. Table 1 shows statistical data for pH and colour.

Figure 3 shows water quality parameters as annual arithmetic means and population standard deviation. In 1983, sulphate was clearly the dominant anion and the buffer capacity was low in most of the localities. Nitrate concentrations were well above background concentrations from remote areas in Norway (Blakar and Hongve, 1997), but still low compared to sulphate. Iron and alu-

**Table 1.** Comparison of water quality data from the 24 lakes in this study with all 56 investigated lakes in the area. Data from October 1983, before the start of liming activities.

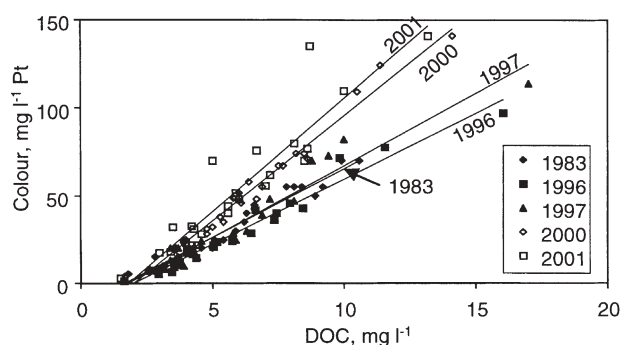
Parameter No. of lakes	pH		Colour (mg $l^{-1}$ Pt)	
	56	24	56	24
Range	4.5–7.2	5.7–7.2	5–70	5–70
Mean	6.1	6.2	32	34
Median	6.2	6.2	25	28
SD	0.48	0.35	20	20



**Figure 3.** Arithmetic mean values and standard deviation for water quality parameters in 24 lakes in the Østmarka area.

minium occurred in enhanced concentrations typical for acidified areas. (Iron concentrations, not shown in Fig. 3, were  $150 \pm 96 \mu\text{g l}^{-1}$ ). In 1996 and 1997 the concentration of sulphate and nitrate were reduced, while the alkalinity was significantly increased ( $p < 0.01$  for both  $\text{HCO}_3^-$  and ANC). However, no significant change in pH was noticed since 1983. Further, DOC, iron and aluminium concentrations were almost unchanged.

In 2000, pH was significantly lower ( $p < 0.01$ ) and up to one unit lower than in previous measurements. Total ion concentrations, measured as electrical conductivity, were reduced too, while aluminium and nitrate concentrations were higher than before. Most parameters, such as pH and ANC did not change significantly from 2000 to 2001. The water colour increased during 1996–2000, although this trend was not accompanied by significant increases in DOC concentrations. As a result, regression lines of colour versus DOC in Figure 4 had slopes about 10:1 for 2000 and 2001 and 8:1 for the other years. Figure 4 shows also that the lake waters can contain around  $2.0 \text{ mg l}^{-1}$  DOC without visible colour.



**Figure 4.** Relationships between water colour and DOC in the studied lakes. Linear regression lines are shown for each year.

Raw water from Skullerud water treatment plant has shown increasing colour during the period 1976–2002 as a total, but the increase has not been continuous (Fig. 5). First, a slight increase in colour occurred during 1976–1988, followed by a decline in 1989–1991, and increasing values since 1992. The steepest increase started in 1998 and continued until the end of 2000. After 2000, the colour declined. Further, the  $\text{COD}_{\text{Mn}}$  and DOC-values have increased since the first part of the 1990s. However, the change that took place by the end of 2000 was more than 40% for colour and only 11–13% for  $\text{COD}_{\text{Mn}}$  and DOC.

A general trend in our data is that solute concentrations varied most in small lakes. This trend is assumed to depend on shorter water retention times for small compared to larger lakes. Figure 6 shows the concentration levels for some selected parameters in three lakes with different estimated water retention times. Lakes with short retention times show declining sulphate concentrations due to reduced concentration in precipitation and inflow water. The same lakes had the most marked increases in colour after 1997.

Field data derived from years that are widely different with respect to precipitation, show that annual mean temperatures were less variable and, except for 1996, warmer than the average for 1961–1990 (Fig. 2). Statistical regression analyses were made for selected water quality parameters versus meteorological (independent) variables. Testing of different methods to express precipitation intensity prior to the sampling events showed that the amount of precipitation during a few months before the sampling occasions gave poor correlations with the physical and chemical variables, while precipitation during approximately 15 months, from September the year before until the end of the sampling month, gave the strongest correlations. For example, water colour versus precipitation was  $r = 0.47$  ( $p > 0.1$ ) for three months and  $r = 0.99$  ( $p = 0.001$ ) for 15 months. Figure 7 shows regression lines for physical and chemical variables versus 15 months' precipitation. Statistically significant correla-

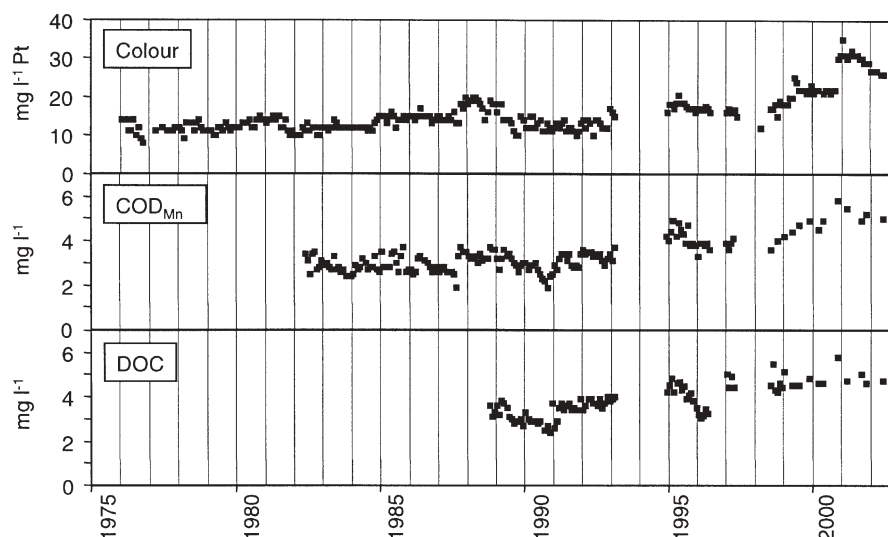


Figure 5. Raw water monitoring results from Skullerud water treatment plant.

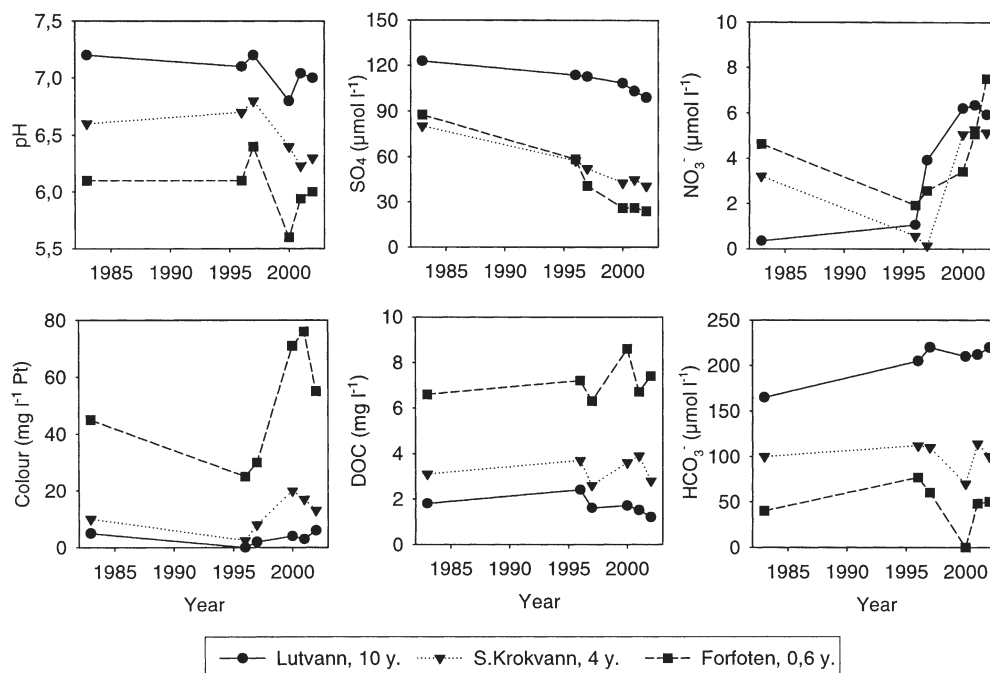


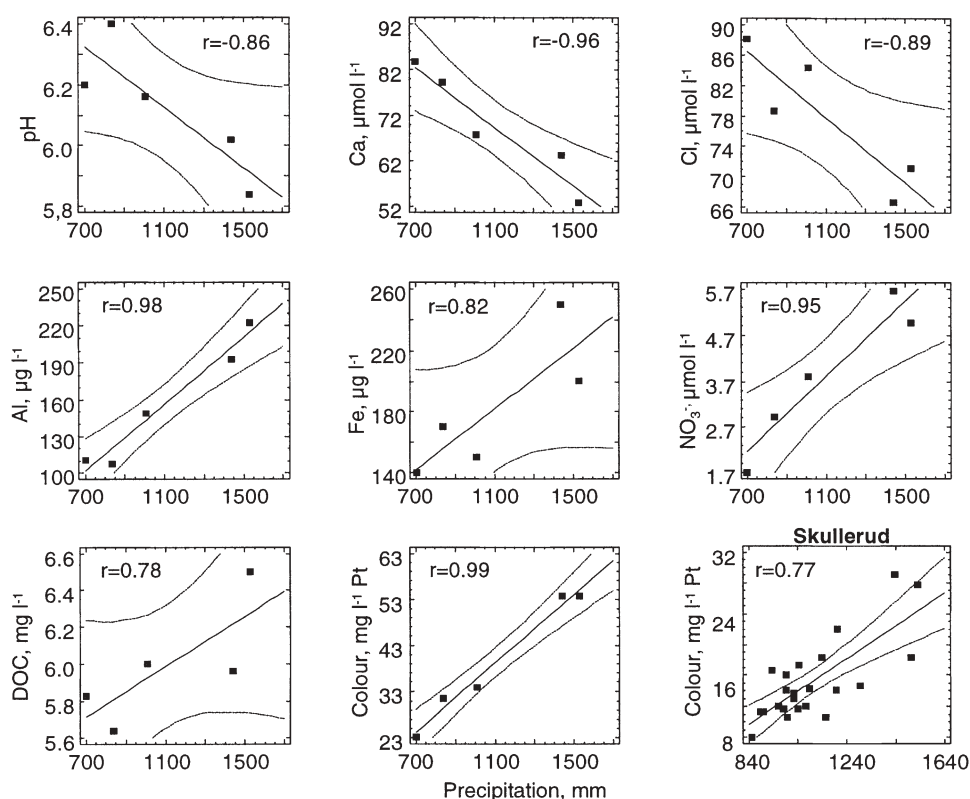
Figure 6. Variations in some water quality parameters in three lakes with different water retention times for the period 1983–2002.

tions are obtained with parameters that express acidification status and colour impact, while the correlation with DOC is not statistically significant ( $p = 0.12$ ). Low flow conditions had less coloured and less acidic discharge than high flow conditions. In contrast, we were unable to see any influence of the annual mean or summer temperatures on water quality.

To test if the connection we found between water colour in lakes and amount of precipitation is valid for the series from Skullerud treatment plant, we extracted mean

values for colour and DOC for October–December each year and made linear regressions with the amount of precipitation in various preceding intervals. Here we found a significant correlation between colour and total precipitation accumulated since September the previous year ( $r = 0.768$ ,  $n = 22$ ,  $p < 0.001$ ) while, the correlation between DOC and precipitation was not statistically significant ( $r = 0.459$ ,  $n = 12$ ,  $p > 0.1$ ). As for the 24 selected lakes, we were unable to see any influence of annual or seasonal temperatures on the Skullerud raw water quality.





**Figure 7.** Physical and chemical water quality parameters versus accumulated precipitation since September the previous year; linear regression lines and 95% confidence limits. The points represent mean values from 24 lakes for 5 different years ( $n = 5$ ). The last graph shows annual values from Skullerud water treatment plant,  $n = 22$ .

## Discussion

Amounts and intensities of precipitation influence the direction of flow through the soil and also the soil water retention time before it enters streams and lakes. Given enough time, reactions with soil minerals neutralise atmospheric acidic deposits and increase the ionic strength of the discharge. Figure 7 shows that not only pH but also  $\text{Cl}^-$ , which originates mostly from precipitation, and  $\text{Ca}^{2+}$ , which comes from dissolution of soil minerals (Gorham, 1961), correlate negatively with the amount of precipitation. Variability in the water quality among lakes is caused by natural differences between catchments with respect to soil thickness, vegetation, extension of mires, etc. The retention time of lake water also is important. Lakes with long retention times respond slowly to changing environmental influences and they maintain a stable water quality due to in-lake processes. Redox- and pH-dependent metals like iron and aluminium settle out of the water column, while sun-bleaching, microbial degradation and coagulation effectively reduce water colour and NOM (Reche et al., 1999).

The anthropogenic acidification process has been ascribed mainly to increased deposition of sulphuric acid. This culminated in the 1970s when successful

measures were taken to reduce  $\text{SO}_2$  emissions. It is therefore noteworthy that the reduction in sulphate concentrations in lake waters between 1983 and 2000 has not led to higher pH. On the other hand, the present data indicate a strong connection between pH and amount of precipitation (Fig. 7). In 1997, a year much dryer than normal, the pH-values increased slightly, while in the wet years 2000 and 2001 the pH values were significantly reduced (Fig. 3). Simultaneously, DOC concentrations were reduced and increased in the dry and the wet years, respectively.

The most notable change during the later years has been in water colour, which has doubled its values. Increases in colour, together with minor changes in DOC, indicate changes in the chemical properties of NOM. Alkalinity, calculated as the difference between sum of base cations and sum of strong acid anions (ANC), were significantly higher ( $p < 0.01$ ) for both 1996/1997 and 2000/2001 compared to 1983, suggesting that no inorganic anions account for the decreased pH in 2000. Another variable that might contribute to decreased pH values is increased values of  $\text{pCO}_2$ . Assuming that the  $\text{pCO}_2$  values during the autumn circulation period are relatively constant from year to year, we suggest that NOM is the major factor that contributed to decreased pH values and

that NOM has become more acidic and coloured as a result of changed water pathways.

Different values for acidity constants and the contribution of NOM to the acidity of surface waters are given in the literature (e.g. Driscoll et al., 1989; Kortelainen, 1993). Our results indicate that these properties of NOM in runoff water can change temporarily due to changes in hydrological flow patterns. Such changing properties of NOM can be explained by well-known properties of NOM components. Hydrophobic acids, which constitute the dominant group of dissolved organic compounds in the humic layer, are effectively retained as water percolates through E and B horizons of podzol profiles (Easthouse et al., 1992). The molecular size of the percolating NOM also is reduced (Riise et al., 2000b). Mobile NOM fractions in the soil are less coloured and absorb less ultraviolet light than hydrophobic acids (Hongve et al., 2000). When high precipitation intensity causes a dominance of flow through the upper soil layers, the concentration of coloured hydrophobic acids in the discharge will increase. It has been observed that high-molecular-weight forms of NOM were most abundant in discharge water during high flow conditions (Riise, 1997), and contributed more to water colour than the low-molecular-weight form of NOM (Hongve, 1989).

## Conclusion

The study showed that concentrations of NOM and water colour have increased in forest lakes in the Østmarka area between 1983 and 2000/2001. In raw water from the Skullerud treatment plant, DOC concentrations and water colour have increased by 50% between 1989/1990 and 2000. Increases in water colour and changes in concentrations of inorganic constituents are well correlated with increasing amounts of precipitation, while no effect is seen from increasing temperature. Changed water pathways, due to increased total precipitation and more periods with very intensive rain, have led to more leaching of coloured and acidic organic compounds from the upper forest floor. This process has increased the colour: DOC ratio and decreased the pH of lake waters. Our results concerning water colour are consistent with former reports of increasing colour in Swedish lakes and streams during the 1970s and 1980s (Forsberg and Petersen, 1990) when annual precipitation was increasing but summer temperatures were generally lower than normal.

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