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Quantifying the Drivers of the Increasing Colored Organic Matter in Boreal Surface Waters

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Long-term monitoring of surface water quality has shown increasing concentrations of colored dissolved organic matter (CDOM) across large parts of the northern latitudes. This has increased purification costs for domestic water works. Appropriate abatement actions require better knowledge of the governing factors for the increase, and this has motivated a growing scientific interest in understanding the factors and mechanisms promoting the CDOM increase. A proposed water color model for an important raw water source for Oslo, Norway, is based on the precipitation's amount and mobile ion concentration. The model explained more than 93% of the temporal variation in CDOM between 1983 and 2008. The model structure was also tested on three adjacent raw water sources and was found to explain 75−82% of the CDOM development throughout the same period. The long-term trend of increasing CDOM was closely related to the decline in sulfate and chloride concentrations in precipitation. Furthermore, interannual fluctuations in CDOM were explained by variation in predominant water flow paths, depending on amounts and intensity of precipitation, both of which are predicted to increase in several parts of the northern latitudes according to climate change scenarios.

Introduction

Concentrations of CDOM in northern European and north-eastern American lakes have increased significantly over the past 2−3 decades (*1*−*5*). SO_2 emissions peaked in northern Europe and North America from the mid 1960s to the mid-1970s (*6*), and in southern Norway there has been a decline in the concentration of acid rain components of about 70% during the past two decades (*7*). In the early 1980s, it was hypothesized that increased inputs of strong mineral acids possibly could explain the decline in CDOM (*8*), which in the previous decades had been observed in several lakes on the northern hemisphere. At present, the increase in CDOM concentrations in northern Europe and North America has been linked to the decreasing acid rain depositions (*1, 9*). Some studies have also pointed out a similar effect caused by sea-salt in precipitation as for the acid rain deposition (*1, 10, 11*). Several authors also mention fluctuations in rainfall amount and intensity, air temperature, and sunlight radiation as regionally important factors, along with the impact of climate change, for the seasonal and interannual variation in CDOM concentration and quality (*12*−*20*). In addition, the importance of catchment characteristics such as soil acidity on CDOM has been recognized (*1, 11*). Catchment runoff chemistry has also been found to be highly influenced by hydrology (*21*). In particular, headwater streams show elevated CDOM concentrations and changes in the quality of CDOM during periods of high runoff (*16*−*20, 22*−*24*). Hence, the predicted changes in precipitation patterns, causing annual flooding in southern Norway (*22*), will further affect the amount and characteristics of CDOM leaching from the soil to surface waters in the future.

The main objective of this study was to assess more directly how temporal trends in CDOM concentrations in boreal lakes of low ionic strength are related to changes in precipitation quality and quantity. This was made possible by long time series of color intensity measurements on raw water for drinking water purposes, conducted by the Oslo Water and Sewerage Works (WSW) where land use within the catchment is heavily restricted, and where the average annual, growing season and winter air temperatures have not changed significantly ($p > 0.05$) throughout the past two decades.

Methods

Study Sites. Four adjacent lakes, which cover the municipal drinking water demand in Oslo, Norway (Figure 1), were included in the study; Elvåga (59°53′N, 10°54′E, 196 m a.s.l.), Maridalsvatnet (59°59′N, 10°47′E, 150 m a.s.l.), Langlivatnet (59°05′N, 10°34′E, 320 m a.s.l.), and Alunsjøen (59°57′N, 10°51′E, 239 m a.s.l.). The catchment bedrock for all lakes

FIGURE 1. Norway and location of the studied raw water sources.

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consists mainly of gneisses and granites, predominantly covered by a thin (typically 0−50 cm), acid-sensitive podzolic soil with scattered mires. The vegetation cover is comprised of low nutrient demanding Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* L.) forest, with a scattered mixture of deciduous trees. Surface waters within the area are generally dilute (conductivity 25−40 μS/cm) and dystrophic. Average annual, growing season (June−August) and winter (December−February) air temperatures have not changed significantly ($p > 0.05$) throughout the past two decades in this area.

Since the studied lakes are drinking water sources, land use within the catchment is heavily restricted. This enabled us to isolate the effect of varying precipitation chemistry and precipitation amounts on the CDOM concentrations in the lakes. Variations in theoretical water retention time and autumn circulation patterns, among the selected lakes, allowed us to test the model for lakes with different settling/resuspension properties with respect to CDOM, which is important for the water color intensity development in the lakes (*25*). Lake surface areas and theoretical retention times are given in Table 1. Since they are raw water sources, the

TABLE 1. Surface Areas and Theoretical Water Retention Times of the Four Studied Lakes

	Elvåga	Maridalsvannet	Langlivatnet	Alunsjø
retention time (yrs)	2	0.27	0.15	1.6
surface area (km^2)	1.39	3.89	0.73	0.39

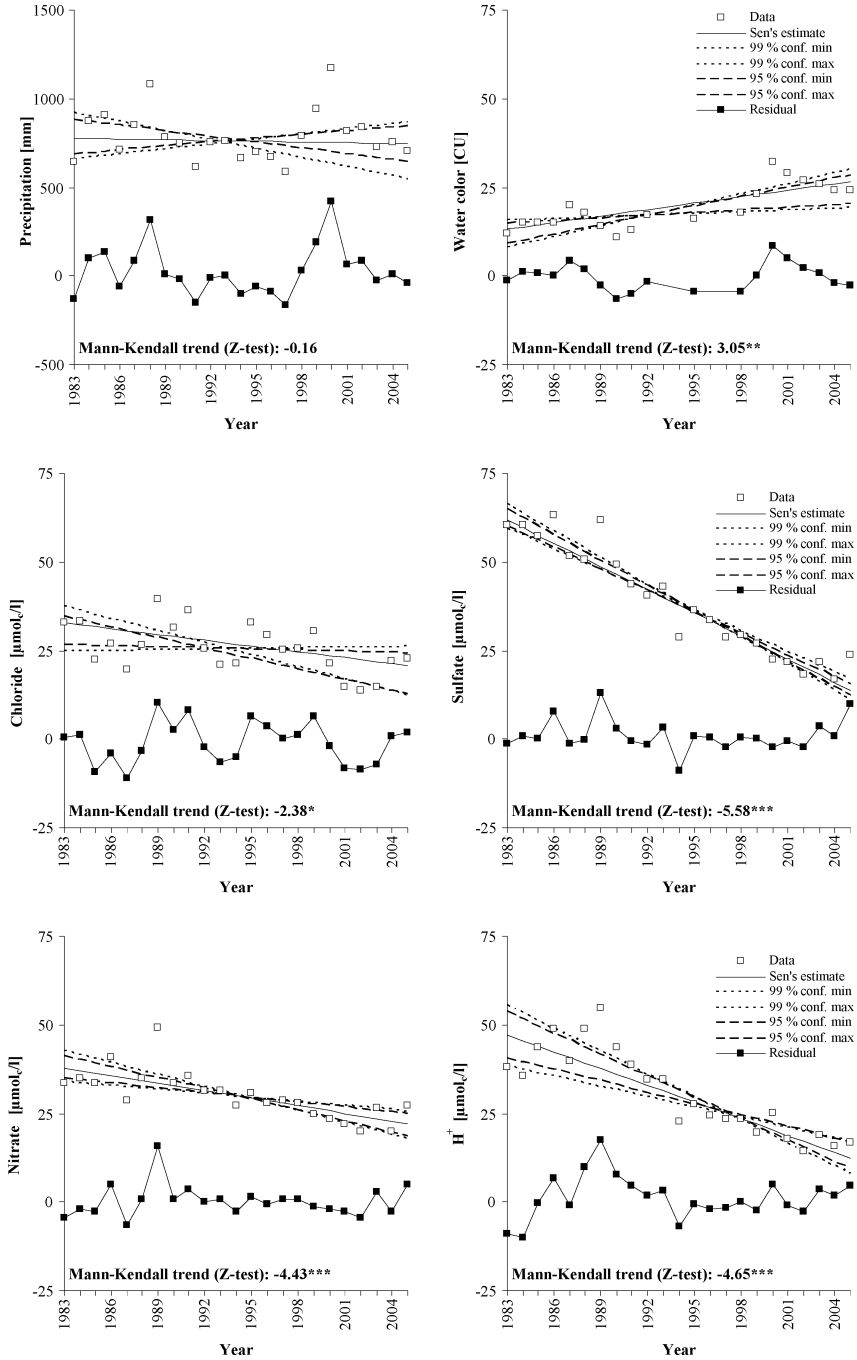

FIGURE 2. Sen's slope residuals and slope estimations for precipitation quality (chloride, sulfate, nitrate, and H$^+$ concentrations), precipitation quantity, and water color intensity in the raw water source Elvåga (1983−2005).

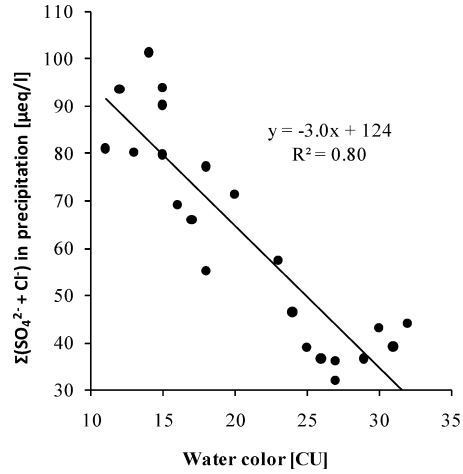

FIGURE 3. The sum of annual average sulfate and chloride concentration ($\mu mol_c/L$) in precipitation vs water color (CU) in December samples from the raw water source Elvåga.

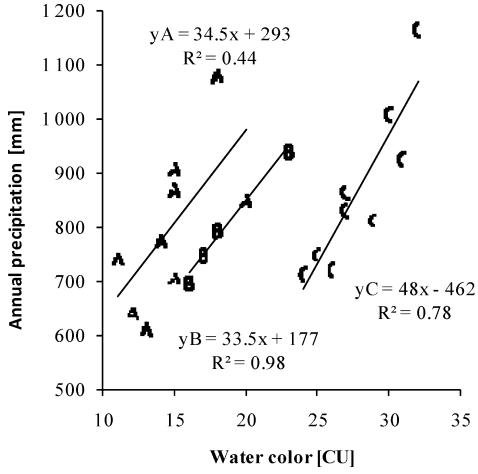

FIGURE 4. Annual precipitation amounts (mm) separated into three quality classes regarding the sum of sulfate and chloride concentration in precipitation ($\mu mol_c/L$) vs water color (CU) in Elvåga. A ≥ 70, B = 50−70, and C ≤ 50.

lakes are all fairly deep, with maximum depths ranging from 30−50 m.

Data Set and Model Approach. The Oslo WSW provided 25 years of monitoring data (1983−2008) from the lake Elvåga on water color intensity, with biweekly to daily resolution. The water was extracted below the thermocline. Corresponding data series (with a monthly resolution) from three adjacent lakes were also provided by the Oslo WSW. The water color intensity was expressed as Hazen color units (CU), where one unit of color is that produced by 1 mg Pt/L, assessed from absorbency at λ410 nm (Abs_{410}) (*26*). Water color intensity can be used as a proxy for CDOM concentration.

Precipitation amounts and chemical composition data were obtained from the Norwegian Meteorological Institute for the nearby (4−14 km) station of Oslo-Blindern and from the Norwegian Institute for Air Research (NILU) at Løken, respectively. Annual long-term mean precipitation for station Oslo-Blindern (1961−1990) is 763 mm.

The Mann-Kendall test for the presence of monotonic long-term trends and a nonparametric Sen's method for estimating the slope for a linear trend for precipitation chemistry and amount were conducted according to Gilbert (*27*), using the MAKESENSE 1.0 software by the Finnish Meteorological Institute (*28*).

Best subset regression analysis, a method used to help determine which predictors should be included in a multiple regression, was used to find the best predictors for the CDOM development in the lake Elvåga. The potential predictors were the annual amounts of precipitation and the annual average concentrations of the major ions in precipitation (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, H^+, SO_4^{2-}, NO_3^-, and Cl^-). This was only done for lake Elvåga since it had the data set with the highest resolution. By using color data from late December, we assured a full circulation within the lake and thereby obtained the best possible integrated value of the year's input of allochthonous organic matter. A multiple linear regression analysis was used to determine the optimal set of predictors, all of which had a significant ($p < 0.05$) explanation ability for the color development in lake Elvåga. This set of predictors was used in a model with the following general structure

$$C_t = C_0 + k\cdot(\Delta \text{ precip quality})^a\cdot(\text{precip quantity})^b$$

where C_t is the modeled water color intensity (CU), C_0 is equal to the water color intensity measured in the first year of the monitoring (CU), k is a coefficient adjusting for differences in denomination and scale (CU/(μmol_c/L·mm)), Δ precip quality denotes annual differences in the precipitation chemistry (μmol_c/L), precip quantity denotes annual precipitation amounts (mm), and a and b are constants to adjust catchment site specific weights between precipitation quality and quantity, respectively.

The model structure was tested on corresponding monthly averages of December data for Maridalsvatnet, Langlivatnet, and Alunsjø. For each lake, the model was empirically fitted by using appropriate C_0 value optimizing the model coefficient k and adjusting the site specific constants a and b.

According to the above model, parameters were set for each lake, and the percentage influence (with a 5% step resolution) from the previous year was found by using an optimization of the correlation coefficient between the modeled and the measured color intensity. As an example, we can assume that a 25% influence from the previous year gives the best correlation between the modeled and the measured color intensity. Hence, if for instance 16 CU is modeled from the present year's precipitation data predictors, and 8 CU is the previous year's water color intensity (measured or modeled), then the adjusted modeled water color would be 0.25 · 8 CU + 0.75 · 16 CU = 14 CU. If then 18 CU is modeled for the following year, then the adjusted modeled water color would be 0.25 · 14 CU + 0.75 · 18 CU = 17 CU, and so on.

TABLE 2. Water Color Model Parameters for the Four Investigated Lakes

	Elvåga	Maridalsvannet	Langlivatnet	Alunsjø
C_0	12	10	22	6
k	9.80×10^{-8}	3.25×10^{-9}	4.15×10^{-9}	2.06×10^{-9}
a	1.3	1.3	1.3	1.3
b	2.0	2.5	2.5	2.5
influence from the previous year	30%	25%	0%	20%

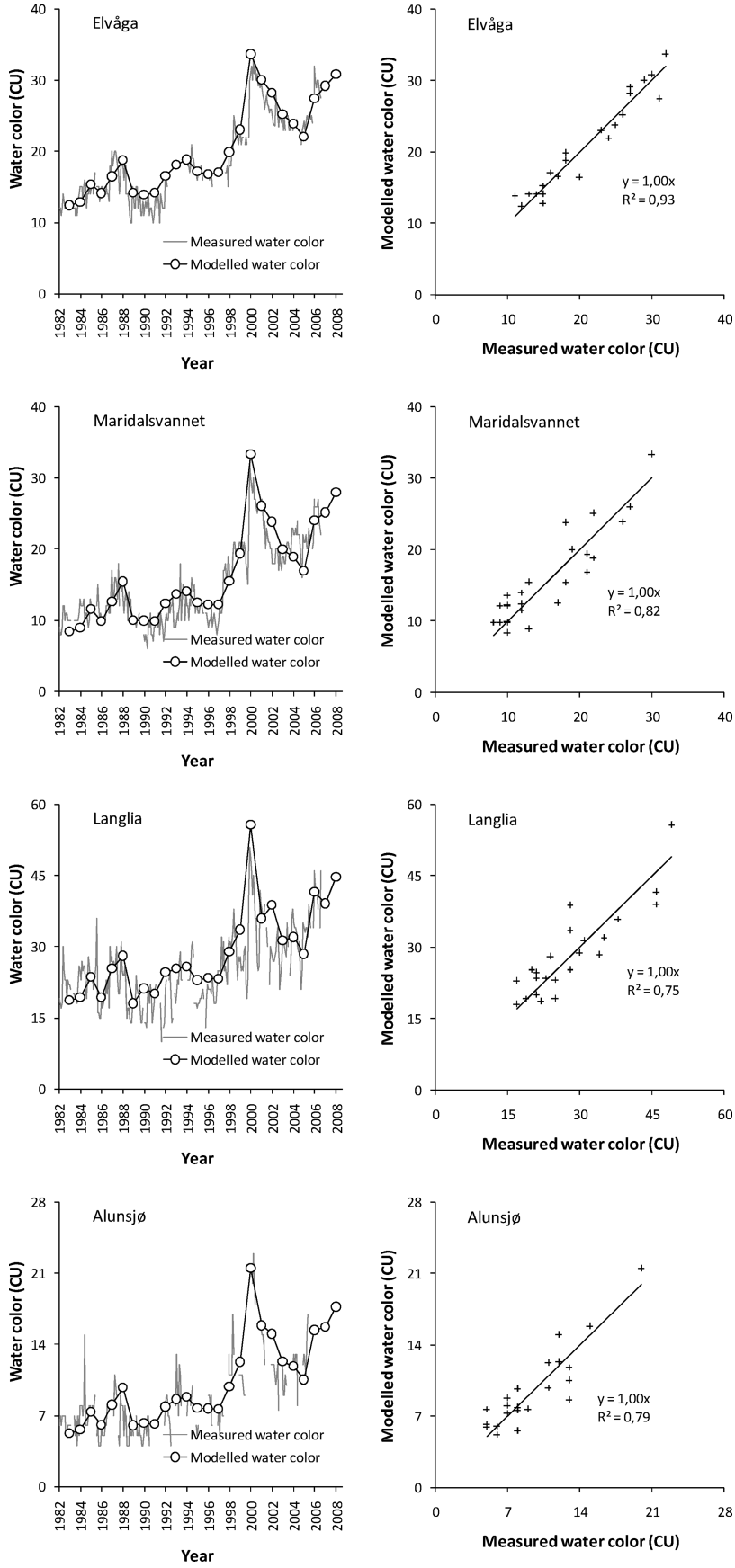

FIGURE 5. Modeled and measured water color for the four raw water sources for Oslo Water and Sewage Works (left column). The water color is for December samples. Linear regression lines for a plot between measured and modeled water color are shown in the right column.

Results

The best set of predictors for the temporal variation in lake water color was found to be the sum of sulfate- and chloride concentrations (i.e., mobile anions) in precipitation, along with the precipitation amounts. The sulfate concentrations in precipitation declined significantly ($p < 0.001$) throughout the study period (Figure 2). Despite substantial interannual variations there was also a significant decline in the chloride concentration ($p < 0.05$) throughout the study period (Figure 2). There were substantial interannual variations in annual amounts of precipitation (531−1172 mm), but no significant time trends ($p > 0.05$) were observed (Figure 2).

The sum of sulfate and chloride concentrations (μmol_c/L) in precipitation explained about 80% of the CDOM development in Elvåga (Figure 3). Chloride would explain some of the interannal variations (Figure 2). The lowest (12 CU) and highest (35 CU) water color intensities were measured in the driest (1990) and wettest (2000) years, respectively, but there was only a weak positive trend between annual precipitation amounts and water color intensity ($r^2 = 0.29$). However, when precipitation was divided into three quality classes, $\Sigma(SO_4^{2-} + Cl^-)$: < 50, 50−70, > 70 (in μmol_c/L), a strong correlation between precipitation amount and water color intensity was found within each quality class (Figure 4). Hence, the annual precipitation amount was interpreted as an overlying explanatory factor in the model, explaining interannual variation in CDOM concentrations.

The fitted model parameters and the percentage correction for the previous year's influence are shown in Table 2. By correcting for previous year's influence, the model for Elvåga explained more than 93% of the CDOM development throughout the study period (Figure 5). When the model structure was refitted by using the monthly average December-data for the three adjacent raw water sources, it explained 75−82% of the CDOM development throughout the same 25-year timespan (Figure 5).

Discussion

Decreased Concentration of Mobile Anions. We believe that the main factor governing trends in water color intensity and increased concentrations of organic matter is decreased ionic strength. This leads to increased surface charge of CDOM species and increased electrostatic repulsion of CDOM and thereby decreased flocculation, coagulation, and precipitation (*29*). In situ studies have pointed out that decreased ionic strength leads to increased export of CDOM (*30*). The ionic strength is especially affected by changes in concentrations of multivalent ions in precipitation, and the decline of sulfate concentrations, along with reduced alumnium (Al^{n+}) leaching, should therefore be of major importance. Flocculation of CDOM using alumnium sulfate as a flocculent is a well-known procedure in water treatment plants. A decrease in inorganic alumnium concentrations in soil solutions is therefore believed to decrease the extent of flocculation and thereby allow for increased export of CDOM from soils to surface waters. Hence, the reduced input of atmospheric acid deposition and sea-salts plays a central role in governing the water color intensity by decreasing the ionic strength of soil solutions and thereby increasing the export of CDOM from catchments to surface waters (*1, 5, 11*). The relative decrease in H^+ and mobile anion concentrations in precipitation (Figure 2) has subsequently reduced the ionic strength and to some extent the H^+ concentrations in lakes, but the increase in pH in lakes in Østmarka has not been as large as could be expected from the reduction of acid deposition (data not shown). Increased concentrations of weak organic humic acids (*31*) and declined concentrations of inorganic alumnium (*32, 33*) and base cations may however counterbalance the observed reduction of mobile anions.

Precipitation Amounts. In three subsets of years with similar mobile anion loading, the water color was found to be correlated to precipitation amounts (Figure 4). Increased amounts of precipitation are inherently related to increased water runoff intensity, especially in regions with shallow soil and impermeable bedrock. Increased runoff intensity reduces the time available for retention processes to occur both in terrestrial and aquatic environments and is known to be associated with increased CDOM concentrations in surface waters through various mechanisms. For instance, increased runoff flux from lakes shortens the CDOM retention time and thereby decreases turnover (*25, 34, 35*) and photooxidation of CDOM (*12, 13, 36, 37*). The differences between the lakes regarding the influence from the previous year can to some extent be explained by the lakes' theoretical water retention time and differing lake and/or catchment characteristics that influence autumn circulation. The lakes Elvåga and Alunsjø have the longest water retention times, while Langlivatnet has the shortest. Maridalsvatnet has a short retention time but by far the largest catchment and lake surface area. A larger lake surface area promotes a better autumn circulation (*38*), and this could plausibly explain the relatively high influence (25%) of precipitated CDOM from the previous year when modeling the water color intensity in Maridalsvatnet (Table 2). Processes upstream of Maridalsvatnet might also influence the prolongation of the previous year's influence. Furthermore, wet years with elevated groundwater tables in catchments with shallow soils promote leaching of more CDOM due to more drainage through upper soil horizons with higher organic matter contents, bypassing the sorption capacity of the deeper mineral soil layers. The upper soil layers have relatively low pH and higher concentrations of CDOM. Such an increased water discharge from a forest floor end-member during periods of high runoff has previously been documented by several episode studies (*39*−*41*). Hence, the plausible conceptual explanation for the overlying interannual variations in color intensity in Lake Elvåga would be increased CDOM leaching from the upper parts of the soil profile during wet years (*42, 43*). Similar hydrological explanations for increased CDOM concentrations in rivers and lakes have also been proposed by others in previous studies within other catchments (*16, 23, 41*).

Future Trends in Leaching of CDOM. The CDOM development in the lake Elvåga is so far primarily explained by the reduction of acid depositions and by natural variations in sea-salt concentrations in precipitation. Annual precipitation amounts explain the strong overlaying interannual variations with higher CDOM concentrations during wetter years. The present precipitation quality leaches more CDOM from catchments to surface waters than precipitation with higher concentrations of acid rain components. Both amounts and intensity of precipitation are suggested to increase according to climate change scenarios (*22*). Hence, despite stabilized atmospheric input of mobile anions, there might still be enhanced leaching of CDOM species from boreal catchment soils in the future.

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