

# More Frequent Runoff and Connected Sources in Headwaters Promote Browning of Northern Freshwaters

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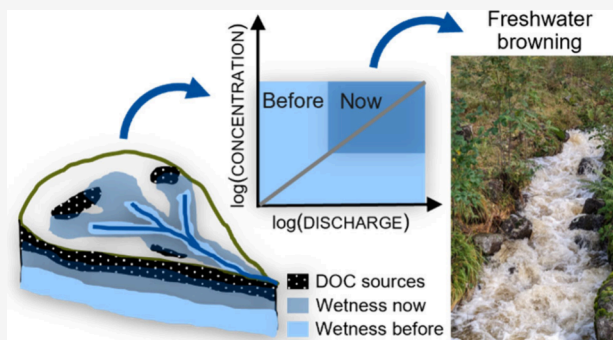
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**ABSTRACT:** Sustained browning of northern waters has prompted inquiries into the drivers of increasing concentrations of organic matter. While reduced sulfur deposition is a key cause, an increasing role of hydrologic mechanisms as a result of cleaner air and progressing climate change has been repeatedly suggested. How these controls act remains however unclear. Here we examine over 30 years of organic carbon concentration and discharge data from four reference streams located across Norway to pinpoint consistent hydrologic changes that may promote water browning. Stable slopes with changing intercepts of the concentration-discharge relations indicate that the influence of air pollution on soil solution chemistry is plausible, supporting available chemical explanations from a hydrologic perspective. Decreasing ratios of concentration to discharge variability, observed in autumn over the years, point to less spatial heterogeneity of the sources of organic carbon. A clear rise in the frequency of runoff events, which increases the opportunities for dissolved organic carbon to transit from soil to streams, also indicates higher hydrologic connectivity and more even mobilization of carbon sources. More connected sources and more frequent runoff events, which jointly enhance the likelihood of organic carbon reaching rivers, may thus contribute to the observed browning of northern waters.

**KEYWORDS:** Water browning, DOM, hydrologic controls, concentration-discharge relationship, runoff frequency, hydrologic connectivity, climate change



## 1. INTRODUCTION

Surface water browning caused by increased dissolved organic matter is widespread across northern ecosystems.<sup>1</sup> Surface waters with high dissolved organic matter absorb more light, reducing its penetration in the water column and thereby primary productivity,<sup>2</sup> as well as increasing the transfer of contaminants.<sup>3,4</sup> Surface water browning also places challenges on drinking water production<sup>5</sup> and is associated with coastal darkening.<sup>6</sup> Furthermore, larger export of dissolved organic matter from soils is likely to reduce the strength of the land carbon sink.<sup>7,8</sup>

A strong driver of increased dissolved organic matter is the decline of acid deposition, in particular sulfur, as shown by statistical analyses of time series of surface water chemistry in seminatural landscapes,<sup>1</sup> experimental field manipulations,<sup>9</sup> laboratory studies,<sup>10</sup> and physical-chemical models.<sup>11</sup> In this context, higher solubility of organic matter under lower ionic strength was proposed as the mechanism explaining browning.<sup>12</sup> Additionally, browning is also sensitive to climatic variations, in particular of rainfall,<sup>13,14</sup> whose importance may be growing as changes in atmospheric chemistry level off.<sup>7</sup>

A mechanistic understanding of the response of dissolved organic matter to changes of climate and the resulting

alteration of hydrologic dynamics within watersheds is however limited. Available modeling and case studies suggest that an increase of dissolved organic matter concentrations is related to more superficial runoff through carbon-rich topsoil occurring during periods of heavy rainfall.<sup>15,16</sup> Nonetheless, this explanation fails to account for antecedent soil moisture conditions, which often control variations in freshwater chemistry,<sup>17</sup> thus limiting our understanding of the interactions between climate change, hydrology and biogeochemical cycling.

In this study, we use data from across a physioclimatic gradient and methods commonly used in the hydrologic sciences to investigate the mechanisms by which a changing hydrology may be contributing to observed increments of dissolved organic matter, thus clarifying additional controls driving the changing water chemistry of northern freshwater

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bodies. In addition to findings on this subject, the paper presents an approach that relies on simple analysis methods and data that are routinely monitored in many countries, thus facilitating its replication in further watersheds.

## 2. METHODS AND MATERIALS

**2.1. Materials.** Data for this study come from four watersheds distributed across Norway, which span a variety of physioclimatic conditions from the southern regions to the northernmost tip of the country along a gradient of atmospheric pollution (Figure S1). They are (from north to south): Dalelv (drainage area: 3.2 km<sup>2</sup>), Langtjern (4.8 km<sup>2</sup>), Storgama (0.6 km<sup>2</sup>) and Birkenes (0.4 km<sup>2</sup>) (Table S1). This is a set of reference acid-sensitive watersheds in natural conditions (i.e., with no anthropogenic disturbances within their boundaries) where the concentration of chemical elements in the river discharge has been monitored for more than 30 years to detect surface water responses to atmospheric deposition in the framework of the Norwegian program on Monitoring of long-range transboundary air pollution - Water chemical effects.<sup>18</sup> The four watersheds have been monitored since the 1980s (Table S1), have small drainage areas, and display marked trends of aquatic concentration of organic carbon in their streams.

We used time series of Total Organic Carbon (TOC) concentrations measured with weekly or biweekly time steps. TOC mostly consists of DOC (for about 90–95%) in these watersheds.<sup>19</sup> Discharge at the outlets where the concentration of chemicals is monitored is obtained from the corresponding discharge measurement stations operated by the Norwegian Water Resources and Energy Directorate (Table S1). Both discharge and water chemistry data are available online and can be downloaded through application programming interfaces (api.nve.no for discharge and vannmiljo.miljodirektoratet.no for water chemistry data).

**2.2. Methods.** We divided the data series of each watershed into seasons (spring: March–May, summer: June–August, autumn: September–November, winter: December–February) to account for seasonal variations of organic carbon dynamics. We then applied rolling windows encompassing ten consecutive years of the same season and shifting one year at a time to the seasonal data series. The width of the rolling windows is chosen to include enough data for obtaining representative estimates of the variables of interest. We finally computed the values of the variables (i.e., mean and standard deviation of TOC concentration, intercept  $a$  and slope  $b$  of the concentration-discharge relation, ratio between the coefficient of variation of the concentration ( $CV_C$ ) and discharge ( $CV_Q$ ), runoff frequency; see details below) from data in each window and used them in the following analyses.

After computing all variables, we standardized their values to allow for comparison among watersheds with different average conditions. Standardized values  $y$  are computed as  $y = (x - \mu_S) / \sigma_S$ , where  $x$  are the original values calculated for each rolling window,  $\mu_S$  is the mean and  $\sigma_S$  the standard deviation of each variable across the windows. Standardized values indicate how distant the original value of the variable is from the sample mean in terms of the number of standard deviations. Positive and negative standardized values respectively indicate values which are higher or lower than the sample mean.

**2.2.1. Concentration–Discharge Relationships.** For each watershed, season and rolling window, we derived intercept  $a$  and slope  $b$  of the concentration-discharge relation<sup>20</sup>  $C = a \cdot Q^b$

as a least-squares regression between TOC concentration ( $C$ ) and discharge ( $Q$ ) in a double logarithmic space. A positive slope  $b$  indicates that the export of solutes is transport-limited, while a negative slope denotes a source limitation.<sup>21</sup>

**2.2.2.  $CV_C/CV_Q$  Ratio.** For each watershed, season, and rolling window, we computed the ratio between the coefficient of variation of TOC concentration ( $CV_C$ ) and discharge ( $CV_Q$ ). The coefficient of variation measures the variability of a quantity in relation to its mean. It is computed as the ratio between the standard deviation  $\sigma$  and the mean  $\mu$  of the quantity. The  $CV_C/CV_Q$  ratio quantifies the variability of  $C$  with respect to the variability of  $Q$ ,<sup>22</sup> thus characterizing the part of concentration variability which is driven by factors other than discharge variability. These other factors are typically related to the availability of the sources of solutes in a watershed.<sup>23</sup> High ratios denote sources with a heterogeneous distribution in space that are temporarily activated with changing discharge, while low ratios indicate a homogeneous distribution of sources.<sup>21,24</sup>

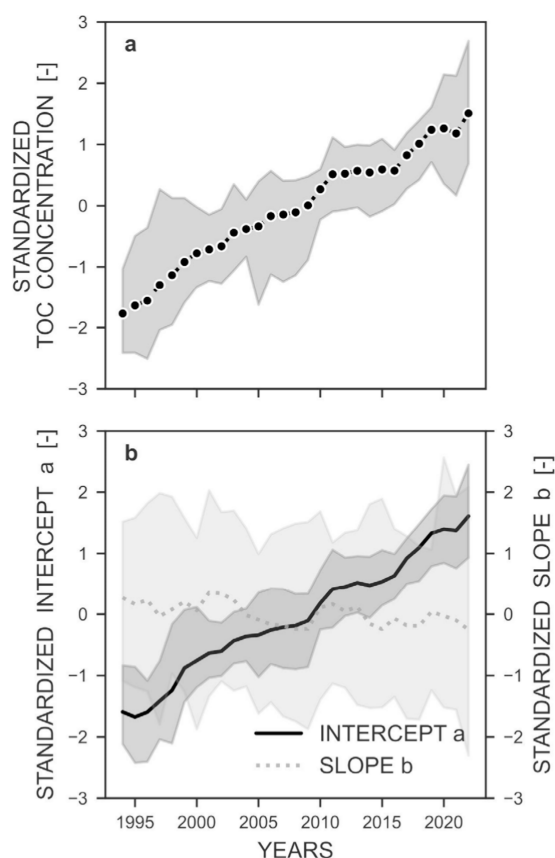
**2.2.3. Runoff Frequency.** For each watershed, season and rolling window, we estimated the frequency of runoff by counting the number of peaks in the discharge series and dividing it by the length of the observations.<sup>25</sup> The *peakdet* function,<sup>26</sup> which identifies local maxima in a vector of data, is used here to count the number of peaks, as it is widely used in the field of hydrology for this purpose<sup>27</sup> and it is available for Python.<sup>28,29</sup> Analogous functions may also be used.

**2.2.4. Trends and Correlations.** We evaluated the existence and magnitude of trends of the standardized variables across consecutive rolling windows by estimating the Theil–Sen slope.<sup>30</sup> This metric is calculated by first computing the mean of all values (e.g., across watersheds and seasons for Figures 1 and 3a) linked to the same rolling window and then estimating the Theil–Sen slope on the basis of these mean values. The result is an overall value of the Theil–Sen slope that accounts for all watersheds and seasons. We computed statistical significance of the trends through a Mann–Kendall trend test modified to account for autocorrelation of the data across rolling windows.<sup>31,32</sup>

We quantified correlations between explanatory (runoff frequency and standard deviation of the TOC concentration) and dependent (mean TOC concentration) variables by means of the Spearman correlation coefficient  $r$ , which provides a nonparametric measure of the strength and direction of an association between two ranked variables. The Spearman correlation coefficient is used to evaluate nonlinear monotonic relationships. It varies between  $-1$  and  $+1$ , assuming these values when one of the variables is a monotonic function of the other.

## 3. RESULTS AND DISCUSSION

Concentrations of TOC in the discharge of the reference rivers steadily increased from the beginning of the data series (Figure 1a; Sen slope = 0.108, Table S2). The increment of TOC concentration is similar for all case studies (Figure S2a), and it is mirrored by TOC fluxes which increase more than discharge (Figure S2b). The phenomenon—known as freshwater browning—is common to most northern watersheds, as many studies before have shown.<sup>1,7</sup> Increasing TOC concentrations can be traced back to steady growths of the intercept  $a$  of the relation between concentration and discharge (Figure 1b; Sen slope = 0.111), whereas only minor modifications of the slope  $b$  of the C–Q relation are observed

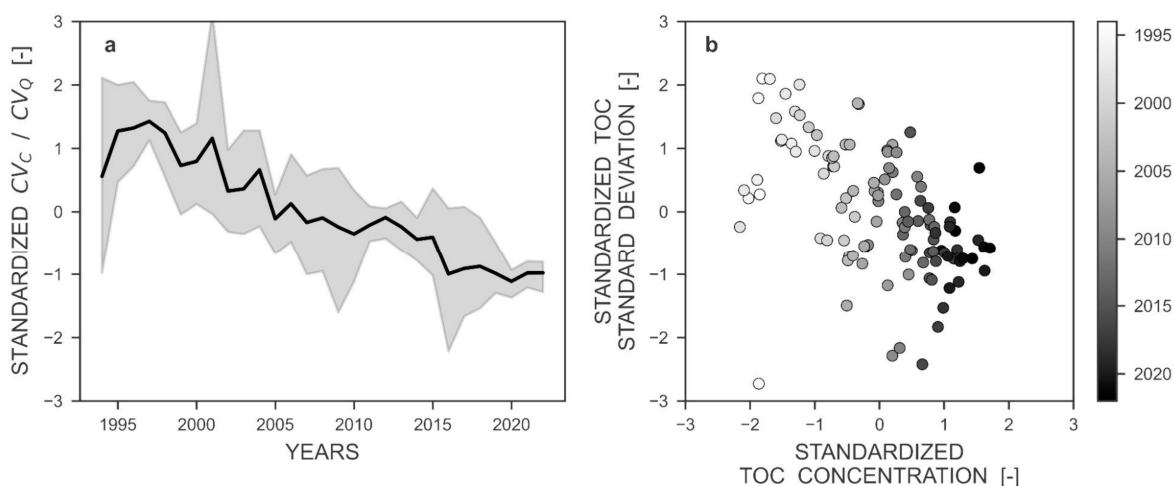


**Figure 1.** Trends in time of (a) standardized mean concentrations of Total Organic Carbon (TOC) and (b) standardized values of intercepts  $a$  and slopes  $b$  of the relations between concentration and discharge for all watersheds and seasons considered in the study. Lines represent mean values and shaded areas the 2.5th–97.5th percentiles of the variability across watersheds and seasons. Years on the  $x$ -axes indicate the last year included in the rolling window used to calculate values of the variables.

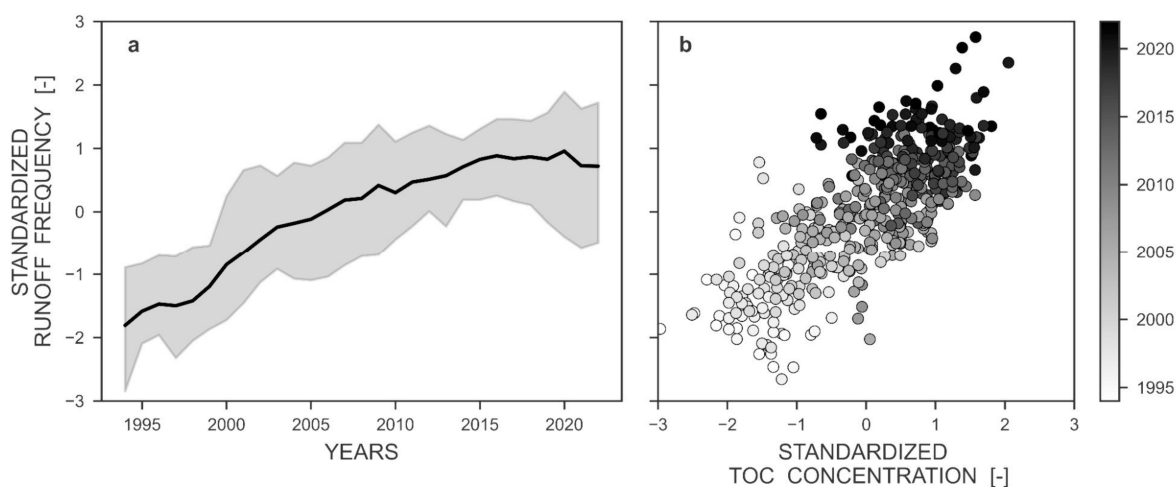
(Figure 1b; Sen slope =  $-0.014$ ). The latter result signals that the rates of change of TOC concentrations with discharge remained stable across the years; otherwise, the slope  $b$  would have changed (absolute slope values are shown in Figure S3). Conversely, stable slopes and increasing intercepts jointly indicate that the amounts of organic carbon transported for each unit of discharge similarly increased for both low and high flows in all the analyzed watersheds and seasons.

A larger part of runoff is typically sourced from more superficial soil layers during high flows.<sup>33</sup> Hence, fairly constant values of the slope  $b$  of the  $C$ – $Q$  relations suggest that the availability of TOC varied similarly in superficial and deeper soil layers. Together with the increments of the intercepts  $a$  of the  $C$ – $Q$  relations, they indicate that the availability of dissolved organic carbon increased similarly in the topsoil and in deeper soil layers. In fact, topsoil and deeper soil layers contribute runoff during high and low flows, respectively. Hence, the detected increase of concentration across the entire range of discharge signals a generalized increase in dissolved organic carbon. In addition, the slope  $b$  would have changed if differential increments occurred across the soil profile. Such a generalized increase agrees with the pervasive influence of air pollution on soil solution chemistry suggested by established studies.<sup>1,12,34</sup> The latter point at reduced percolation of sulfate from the soil surface because of less acid deposition as the likely mechanism responsible for the larger availability of organic carbon across the soil profile that eventually leads to higher TOC concentrations and freshwater browning. Although in this work we do not aim at further substantiating those studies, the stable slopes and increasing intercepts of the  $C$ – $Q$  relations identified by our analyses jointly indicate that mechanisms entailing a generalized increase of dissolved organic carbon across the soil profile (such as, e.g., the chemical effects of reduced sulfate percolation) as opposed to mechanisms involving an increase only in the superficial soil layers are plausible.

Recent studies also suggest an increasing importance of climatic and hydrologic controls on TOC trends observed in



**Figure 2.** (a) Trends in time of the standardized ratios  $CV_c/CV_Q$  between the coefficient of variations of TOC concentration,  $CV_c$ , and discharge,  $CV_Q$  during the autumn season (September–November) for all watersheds considered in the study. Lines represent mean values and shaded areas the 2.5–97.5% of the variability across watersheds. (b) Relation between standardized mean concentrations of Total Organic Carbon (TOC) and standard deviation of TOC concentration in the autumn season for all watersheds considered in the study (Spearman  $r = -0.57$ ,  $p = 5e-11$ ). Hues of the markers of panel b and labels on the  $x$ -axes of panel a indicate the last year included in the rolling window used to calculate the values of the variables.



**Figure 3.** (a) Trend in time of the standardized frequency of runoff events for all watersheds and seasons considered in the study. Lines represent mean values and shaded areas the 2.5th–97.5th percentiles of the variability across watersheds and seasons. (b) Relation between standardized mean concentrations of Total Organic Carbon (TOC) and runoff frequency for all watersheds and seasons considered in the study (Spearman  $r = 0.74$ ,  $p = 2e-78$ ). Hues of the markers of panel b and labels on the  $x$ -axes of panel a indicate the last year included in the rolling window used to calculate values of the variables.

northern watersheds as a result of stabilizing levels of acid depositions.<sup>7,12</sup> How these referenced hydrologic controls act is, however, still unclear. To better understand the mechanisms at work, we analyzed the ratio<sup>22</sup> between the coefficient of variation of the concentration,  $CV_C$ , and discharge,  $CV_Q$ . The ratio  $CV_C/CV_Q$  allows for tracking the portion of concentration variability which originates from other factors than discharge variability, chiefly the spatial variability of the sources of solutes in a watershed.<sup>23</sup>

No clear trends in the ratios were identified in spring, summer, and winter (Figure S4). In the autumn season, when the largest TOC concentrations and fluxes occur on average,  $CV_C/CV_Q$  ratios instead decreased over the years in all of the analyzed watersheds (Figure 2a) despite decreasing discharge variability. The trends of  $CV_C/CV_Q$  ratios in autumn are characterized by a Sen slope equal to  $-0.091$ , indicating a 9% reduction of  $CV_C/CV_Q$  ratios between adjacent 10-years-long time windows. Low values of the  $CV_C/CV_Q$  ratios typically indicate watersheds where solutes are readily available for transport. Wide availability of solutes indeed produces a smoothing of the effect of discharge variability on concentration, thus resulting in lower concentration variability.<sup>21,24</sup> The lowering of  $CV_C/CV_Q$  ratios in autumn may thus suggest that sources of dissolved organic carbon produced by the degradation of soil organic matter during the preceding warm season are becoming more available for transport in these watersheds.

Declining  $CV_C/CV_Q$  ratios are also often considered indicative of decreasing spatial heterogeneity of solute sources.<sup>23</sup> The trend of Figure 2a may thus signal that organic carbon is supplied more homogeneously from source areas in the analyzed watersheds. The effects of such a change in the contribution of organic carbon from the source areas are emphasized by the results of Figure 2b: decreasing variability of TOC concentration (quantified through its standard deviation) along the years (gray shades) correlates well with growing concentration of aquatic organic carbon (Spearman  $r = -0.57$ ,  $p = 5e-11$ ), which causes freshwater browning.

The identified increased homogeneity of the sources of organic carbon may reflect sources that are more evenly

connected to streams than before, as a result of greater hydrologic connectivity. The latter may increase as a consequence of, e.g., increased wetness of watersheds.<sup>35</sup> Direct observations of the wetness are not available for the analyzed watersheds. Hence, we investigated this hypothesis by estimating the frequency of runoff events, which is typically higher in wetter watersheds because of high soil moisture levels, regardless of them resulting from more frequent rainfall or less evapotranspiration.<sup>36</sup>

Findings in Figure 3 support a generalized increase of hydrologic connectivity in the watersheds. In fact, the runoff frequency steadily grew along the years (Figure 3a) with trends characterized by a Sen slope equal to 0.096 (Table S2), i.e., a 10% increase between adjacent 10-years-long time windows. In the autumn season (Figure 2), higher hydrologic connectivity homogenizes the contribution from source areas of widely available organic carbon produced in the warm season (despite a more limited growth of the runoff frequency; Table S2). The effect of such an increase extends beyond making organic carbon sources more connected to streams though. In fact, the runoff frequency grows in all watersheds and seasons and strongly correlates with the observed increase of organic carbon concentration (Figure 3b; Spearman  $r = 0.74$ ,  $p = 2e-78$ ). The result suggests that ongoing hydro-climatic changes such as the observed increment of precipitation<sup>37</sup> (Figure S5), which are consistently increasing the hydrological connectivity of watersheds, also cause more frequent runoff events and thus more opportunities for the organic carbon dissolved in soil water to be delivered to streams.

In summary, more hydrologically connected sources jointly with more frequent runoff events, which increase the chances of solutes being delivered to streams, contribute to the observed rise of organic carbon concentration in northern freshwaters. These two mechanisms are determined by ongoing modifications of the hydrology of northern regions caused by increasingly wetter watershed conditions resulting from a changing climate.<sup>35,38</sup> Our findings also imply that a relatively larger fraction of the decomposition products of soil organic matter is transported from soils to water. All else being equal, such an increasing aquatic export of terrigenous organic

matter represents a lowering of the soil carbon sink strength, as also suggested by simulations of process-based land-surface models.<sup>8</sup>

The present results were obtained from data of four representative watersheds located along a south-north transect spanning the entire length of Norway. Although these watersheds encompass a variety of physioclimatic conditions and analogous conclusions were recently obtained from a study of a large data set of lakes in Canada,<sup>39</sup> we acknowledge possible limitations arising from the small number of case studies. In particular, hydrologic controls of trends of aquatic organic carbon may differ for larger watersheds experiencing weaker increments and for which the relevance of shallow water pathways diminishes.<sup>40</sup> Nonetheless, the key importance of considering small watersheds in the study of dissolved organic carbon fluxes is increasingly recognized.<sup>41</sup> Moreover, the types of data (i.e., daily discharge and low frequency TOC concentration, which are routinely monitored in many countries<sup>42</sup>) and the simplicity of the analyses used in this work facilitate their replication in further watersheds for which data may be available to fellow researchers. This will enable us to ascertain the degree of generality of the identified hydrologic mechanisms contributing to freshwater browning.

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.4c00939>.

Additional details about data series used in the study, observed trends, and slopes of concentration-discharge relationships (PDF)

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### Notes

The authors declare no competing financial interest.

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