Water Research 203 (2021) 117544

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Brownification on hold: What traditional analyses miss in extended surface water records

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ARTICLE INFO

Keywords:
Brownification
Organic matter
Absorbance
Watercourses
Long-term trends
Generalized additive mixed model

ABSTRACT

Widespread increases in organic matter (OM) content of surface waters, as measured by color and organic carbon (OC), are a major issue for aquatic ecosystems. Long-term monitoring programs revealed the issue of “brownification”, with climate change, land cover changes and recovery from acidification all suspected to be major drivers or contributing factors. While many studies have focused on the impact and drivers, fewer have followed up on whether brownification is continuing. As time-series of OM data lengthen, conventional data-analysis approaches miss important information on when changes occur. To better identify temporal OM patterns during three decades (1990–2020) of systematic monitoring, we used generalized additive models to analyze 164 time-series from watercourses located across Sweden. Increases in OC that were widespread during 1990–2010 ceased a decade ago, and most color increases ceased 20 years ago. These findings highlight the need to reassess the understanding of brownification’s spatial and temporal extent, as well as the tools used to analyze lengthening time series.

1. Introduction

In early 2000, trends of increasing organic matter (OM), measured as organic carbon (OC) concentrations and/or water color, were reported in monitoring records from many European and North American surface waters (de Wit et al., 2007; Driscoll et al., 2003; Evans et al., 2005; Monteith et al., 2007; Roulet and Moore, 2006; Worrall et al., 2003; Worrall et al., 2004). The starting points for these increases varied from the 1960 s (Worrall et al., 2003; Worrall et al., 2004) to 1980 s/1990 s (Driscoll et al., 2003; Evans et al., 2006; Evans et al., 2005; Monteith et al., 2007). Since then, there have been predictions that this OM increase, often referred to as brownification, will continue (de Wit et al., 2016; Kritzberg, 2017; Škerlep et al., 2020; Weyhenmeyer et al., 2012). Numerous studies have pointed out the serious ecological and water quality implications of this brownification, such as changes in aquatic food web structures, nutrient availability, pH and metal speciation as well as thermal stratification (Creed et al., 2018; Karlsson et al., 2009; Nova et al., 2019). Elevated OM concentrations can also have negative effects on drinking water treatment (Bieroza et al., 2009; Krasner et al., 1989; Rook, 1974), mobilize pollutants (Bishop et al., 2020; Creed et al., 2018; Laudon et al., 2021) and increase aquatic emissions of greenhouse gases (Lapierre et al., 2013).

Studies investigating the causes of increasing OM concentrations have identified a number of drivers. Declines in acid deposition after it peaked in the 1980 s have been a common explanation (Evans et al., 2005; Lawrence and Roy, 2021; Monteith et al., 2007; Redden et al., 2021; SanClements et al., 2012). Other studies suggested that additional factors related to climate change, land use and biomass increases also contributed to brownification of many surface waters (de Wit et al., 2016; Erlandsson et al., 2008; Finstad et al., 2016; Kritzberg, 2017; Meyer-Jacob et al., 2019; Škerlep et al., 2020; Weyhenmeyer et al., 2012; Weyhenmeyer and Karlsson, 2009). If recovery from acidification is the main driver for the observed trends in OM that would imply that the increasing trends would end as acidification recovery culminates. Other drivers will be continuing (such as changing climate), or at least have no clearly demarcated endpoint (such as biomass increases or land use) (Bragée et al., 2015).

Many of the initial publications using OC as a proxy for surface water

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https://doi.org/10.1016/j.watres.2021.117544
Received 7 June 2021; Received in revised form 6 August 2021; Accepted 7 August 2021

OM were based on a decade, or at most two decades of direct observations. Longer-term studies relied on other proxies such as water color (e.g. Kritzberg, 2017), or paleo-ecological studies (e.g. Bragée et al. 2015, Meyer-Jacob et al. 2019). Surface water monitoring programs have been continuing so now three decades of systematic OC concentration and quality measures (e.g. various absorbance metrics) are becoming available. Hitherto, though, few studies have followed up on whether areas where geographically widespread brownification has previously been reported are continuing to “brownify”. Those few follow-ups, from Finland (Lepistö et al., 2021; Räike et al., 2016), northeastern US (Lapierre et al., 2021) and Nova Scotia (Redden et al., 2021), have found continued browning in a majority of the sites studied.

As data records lengthen, the challenge of analyzing the records is also increasing. A commonly used statistical method to test for temporal trends in environmental data are non-parametric Mann-Kendall tests, since these tests are robust, can easily handle outliers and only require trends to be monotonic but not linear. Such Mann-Kendall tests were also used in many of the above-cited research papers to determine if OM trends were present during pre-defined time periods (Meyer-Jacob et al., 2019; Räike et al., 2016; Worrall et al., 2020). This is, however, an approach that does not retain any information about the exact shape of the trend and its results can be dependent on the time periods chosen for analysis. To improve the evaluation, refined statistical methods, using smoothing methods, like generalized additive mixed models (GAMM) (Hastie and Tibshirani, 1986; Wood, 2017) have been increasingly used to model temporal trends for long time series. These do not require a priori assumptions about the shape of the trends and allow for evaluation of when changes have occurred during the study period.

The aim of this study was to determine whether the brownification previously observed is continuing by evaluating three decades of OM data from 164 watercourses across Sweden using GAMM. The catchments, all with minimal influence from urban or point-source pollution, covered a latitudinal extent of 1600 km. A range of land cover, historic acid deposition levels, and climatic zones from temperate to boreal and arctic, made these data well suited to assessing how extensive brownification remains in space and time.

2. Methods

2.1. Data selection

This study included water chemistry data from 164 watercourses within the Swedish national monitoring program (Miljödata-MVM, 2021; Fölster et al., 2014). The sampling locations extended across 13° of latitude (55 ° 27′ to 68 ° 21′). Catchment sizes ranged from 11 km^2 (10th percentile) to 11,532 km^2 (90th percentile), with a median size of 214 km^2 (Table S1). The mean annual precipitation for each site ranged from 450 mm in the north-east to 1235 mm in the south-west of Sweden. Mean annual temperature ranged from -1.9 °C in the north to 8.9 °C in the south of Sweden. The watercourses were sampled for up to 31 years, between January 1990 and December 2020, for total organic carbon (TOC) and absorbance at a wave-length of 420 nm, hereafter referred to as colored dissolved organic matter (CDOM). Of these watercourses, 65 (TOC) and 101 (CDOM) were sampled in 1990 and 156 (TOC and CDOM) in 2020, with 27 (TOC) and 28 (CDOM) years being the average consecutive duration of each time series. The sampling frequency was monthly for a majority of sites ($n = 117$), up to twice a month in some sites ($n = 37$) and down to bi-monthly in a smaller set of sites ($n = 10$).

All the chemical analyses were carried out at the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (https://www.slu.se/en/departments/aquatic-sciences-assessment/). The field and laboratory methodologies are standardized and well documented. The fact that the same laboratory has analyzed the samples over the entire period (1990–2020) has led to a high level of consistency. In the current study TOC concentration and CDOM data were evaluated. The laboratory is certified (SWEDAC) for analyses of TOC and CDOM. The TOC was measured by combustion methods following standard methods (SS-EN 1484). The absorbance at 420 nm was measured on filtered water with spectrophotometric methods (SS-EN ISO 7887–2012). Organic carbon was measured as TOC which contains $>$ 90% DOC (Humborg et al., 2010). So while the organic carbon concentrations reported are TOC, these can be considered equivalent to DOC. The ratio between CDOM and TOC, the specific visible spectrum (VIS) absorbance (sVISa), was calculated as a measure of OM quality, with a high sVISa indicating more colored OM.

2.2. Statistical analyses

Analysis of station-wise trends was conducted using generalized additive mixed models (GAMM) (Hastie and Tibshirani, 1986; Wood, 2017). This approach allows the modeling of environmental timeseries without prior definition of the shape of the trend curve. The time trend in TOC, CDOM and sVISa, were analyzed using a thin plate spline, while the seasonal component was modeled by a cyclic cubic regression spline with an annual period. As observations must be assumed to be dependent in time, the error term includes a continuous autoregressive process of lag 1 (AR(1)).

First derivatives of the smoothed trend and the corresponding confidence bands were used to determine if a trend was significant at any given time point (Monteith et al., 2014; Simpson, 2018). The obtained information was summarized and visualized according to the suggestions in von Brömssen et al. (2021).

3. Results and discussion

3.1. Temporal trends in organic carbon concentrations

The observed trends in TOC concentrations and CDOM over the last three decades were neither linear, nor monotonic across the data set of Swedish watercourses. The GAMM-based trend analyses indicated that a majority of the sites increased significantly in TOC concentration for some parts of the period 1990–2010 (Fig. S1). There was also a clear pattern of stronger increases in TOC during two specific time periods of 3–5 years each, centered around 1996 and 2007 (Fig. 1, Fig. S1). During the latter peak (2005–2008), up to 50% of the watercourses had increasing trends in TOC. While the trends were not uniform over the period 1990–2010 (Fig. S1), the GAMM analyses detected a breakpoint at about 2010. After 2010, increasing trends of TOC were detected in less than 20% of the watercourses (Fig. 1). This means that while TOC concentrations increased in many areas between 1990 and 2010, the TOC concentrations have been more stable during the past decade (Fig. 3). The cessation of increases in CDOM is just as abrupt, but came even earlier. The trends in CDOM followed that of TOC quite well up to 2000, but then flattened when less than 20% of the sites had increasing CDOM trends (Fig. 1). The share of sites with increasing CDOM trends have gradually decreased since 2000. After 2010, less than 10% of the sites had increasing trends, and approximately the same amount of sites had decreasing trends for a shorter period of time (around 2008–2017). Thus, brownification either has ceased across much of the country, or has at least been put “on hold”.

Temporal patterns in TOC concentrations from eight example sites (Fig. 2) illustrated the trend characteristics for a majority of all sites. TOC increases were present during distinct time periods or during the whole period up to 2010, and then flatten out (Fig. 2c–e) or declined (Fig. 2a). Most of the sites that still show an increase in TOC after 2010 were identified by the GAMM analyses as linear trends (see for example Fig. 2b). This means that for these sites there has been a general increase in TOC over the whole time period and not specifically during the period after 2010.

Appropriate statistical methods need to be used when such complex time trends are described. While the analysis using GAMM allowed us to describe how TOC trends changed during different periods, more

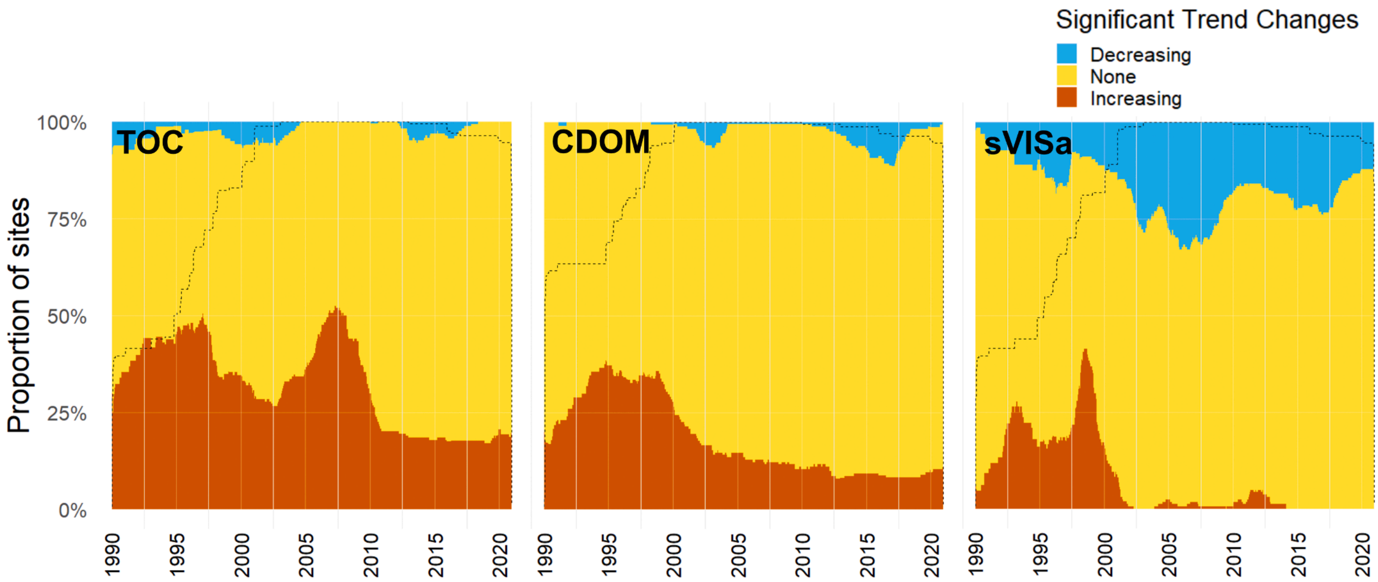

Fig. 1. The proportion of watercourses showing a significant increasing, decreasing or no trend in total organic carbon (TOC) concentration (left), colored dissolved organic matter(CDOM) (center), and specific VIS absorbency (sVISa) (right) during the period 1990–2020. Trends were analyzed by generalized additive models (GAMM). The dashed lines show the proportion of the total number of stations with data for that time point.

commonly used methods, such as the Mann-Kendall test, summarize trends over the entire available time period. Applied to our data, such an approach would have shown that most of the TOC series, and a large proportion of CDOM series, have had an upward trend (119 for TOC and 70 for CDOM out of the total of 164 series). Such an analysis would correctly identify the watercourses where TOC or CDOM had increased, but fail to identify when the increases occurred. As a consequence, such results might lead to the misconception that brownification was still ongoing.

3.2. *Temporal trends in organic carbon quality*

The fact that the trends of TOC and CDOM differed indicated a shift in OM quality over time. The characteristics of OM, as indicated by CDOM normalized by TOC (sVISa), is a tantalizing clue that may also provide some insight into the factors changing surface water carbon. From 1990 to 2000, there were more sites with increasing sVISa than sites with decreasing trends (Fig. 1). But since 2000, sVISa has decreased in around 10–30% of the sites, with almost no sites showing increasing trends. A shift in surface water OM quality over time has been detected in earlier studies of UK catchments influenced by urban areas (Worrall and Burt, 2010). However, those temporal patterns differed from the patterns found in the catchments of the present study where urban areas had very limited influence, if any. A decoupling of DOC and color was also identified in lakes of northeastern US (Lapierre et al., 2021) and Norway (Hongve et al., 2004). This could indicate different drivers for the colored fraction of OM. The OM quality results also demonstrate that trends of brownification are not straightforward, and depend on whether it is color or OC that is considered.

3.3. *Potential drivers for OM trends*

Multiple drivers may have contributed to the rising TOC concentrations prior to 2010 when such trends were more widespread. Hence, it is challenging to distinguish the relative importance of causes for the TOC trends, or lack thereof, observed during the past decade. A systematic investigation of the available evidence to answer this, such as the full range of chemical constituents besides TOC, as well as changes in catchment land use and biomass, soils and climate patterns were outside the scope of this paper that aims to assess the duration and extent of brownification in Sweden. Nonetheless, the spatial and temporal patterns of TOC concentrations across three decades can be used to speculate on the potential drivers.

The fact that brownification has been put on hold since 2010 in most areas of Sweden, could imply that acidification recovery has been one of the major factors driving the trends in TOC after 1990. It could also be that certain climate factors (such as seasonal variation in precipitation and temperature) may be driving trends that are less monotonic in time and space than recovery from acid deposition (Clark et al., 2010). Increasing biomass and/or land cover changes is a persistent factor that could drive brownification in the more limited areas where increases in TOC continued after 2010. Indeed, many of the sites that have continued to brownify are located in the south of Sweden where poor agricultural land was converted to coniferous forest a little over a century ago (Lindbladh et al., 2014), while coniferous forests have been widespread in other parts of the country for many centuries. The increase in conifer forests, specifically spruce, has been suggested as one driver for recent (i.e. post 1900) brownification (Kritzberg, 2017; Škerlep et al., 2020) in southern Sweden. The appearance and loss of spruce in the landscape has also been linked to browning and debrowning of Swedish lakes in earlier millenia (Meyer-Jacob et al., 2015). Southern Sweden is however also one of the areas in Sweden that received the most acid deposition and may therefore be taking longer to recover from acidification. Another region where TOC has increased after 2010 is the north-east coast of Sweden. This coastal zone is distinguished from the rest of the country as an area where isostatic rebound after the last glaciation continues to raise the land out of the Baltic Sea (Tuittila et al., 2013). The relatively flat topography of the new land emerging from the ocean has created shallow peatlands that sustain high amounts of OM in surface waters (Ivarsson and Jansson, 1994).

4. Conclusion

The key finding of our study is that significant increases in TOC concentrations have been absent for a decade over much of a region previously identified as undergoing a strong and persistent brownification. While this is just one of several geographical regions where widespread OM increases were earlier identified, it challenges the impression that these increases are still ongoing. It also calls into question predictions that ongoing pressures such as climate warming or

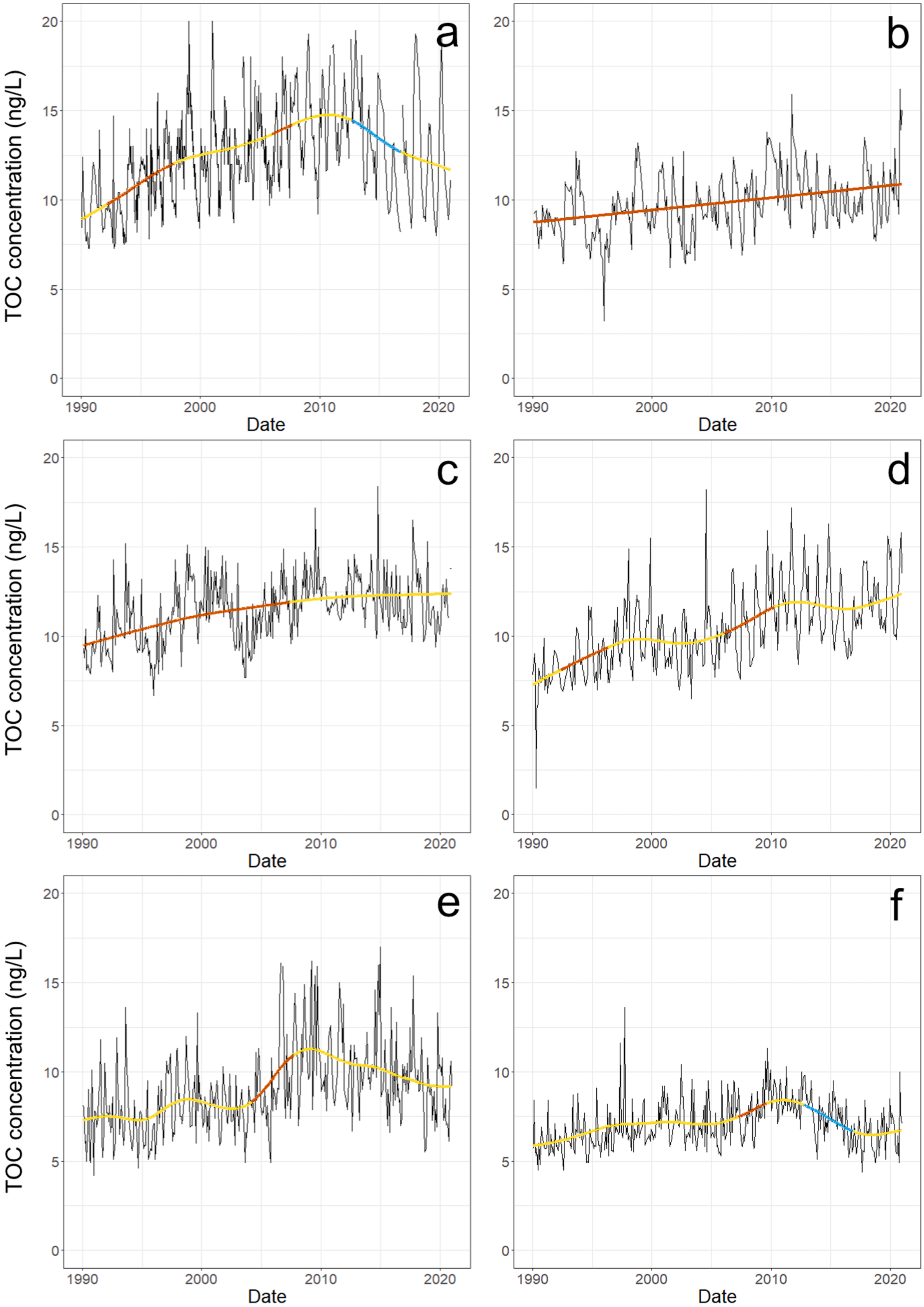

Fig. 2. Examples of time series and trend curves for TOC concentration data between 1990 and 2020 from six of the watercourses included in this study, (a) Emån (site #226), (b) Gide älv (site #301), (c) Rickleån (site #456), (d) Örekilsälven (site #478), (e) Skivarpsån (site #566), and (f) Dalälven (site #326). The smooth curve is produced by a generalized additive mixed model (GAMM). Reddish-orange color indicates periods with increasing trends, blue indicates periods with decreasing trends, and yellow indicates no significant trend.

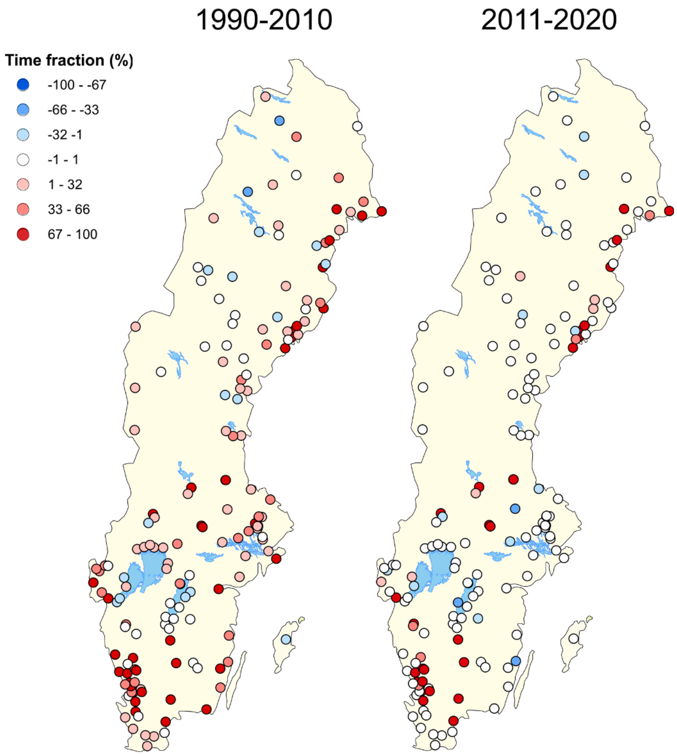

Fig. 3. The time fraction (%) of significantly increasing or decreasing trends before 2010 and after 2010 (left and right panels, respectively). Red colors indicate increasing trends, blue colors indicate decreasing trends, and white color indicate no significant trend. If there are both increasing and decreasing trends occurring during each period, one month of increasing trends canceled out one month of decreasing trends.

increasing atmospheric CO_2 are driving widespread brownification. The lengthening of observational records, together with the geographical variation in the duration and direction of trends across Sweden provide new opportunities to test hypotheses about factors controlling brownification. Above all, this study is a reminder of the need to continue with monitoring and to use appropriate statistical methodology to reconsider earlier findings, especially when those findings are the basis for steering far-reaching management initiatives.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This study was funded by the Swedish Environmental Protection Agency (Project Number NV-02868-18) and the Swedish University of Agricultural Sciences environmental monitoring and assessment programme “Lake and watercourses”. We greatly appreciate the friendly review of this manuscript by Christopher D. Evans.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2021.117544.

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