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Current Browning of Surface Waters Will Be Further Promoted by Wetter Climate

Heleen A. de Wit,[*,†] Salar Valinia,[†] Gesa A. Weyhenmeyer,[‡] Martyn N. Futter,[§] Pirkko Kortelainen,[∥] Kari Austnes,[†] Dag O. Hessen,[⊥] Antti Räike,[∥] Hjalmar Laudon,[#] and Jussi Vuorenmaa[∥]

[†]Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, 0349 Oslo, Norway

[‡]Department of Ecology and Genetics/Limnology, Uppsala University, Norbyvägen 18D, 752 36 Uppsala, Sweden

[§]Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, Lennart Hjälms väg 9, 750 07 Uppsala, Sweden

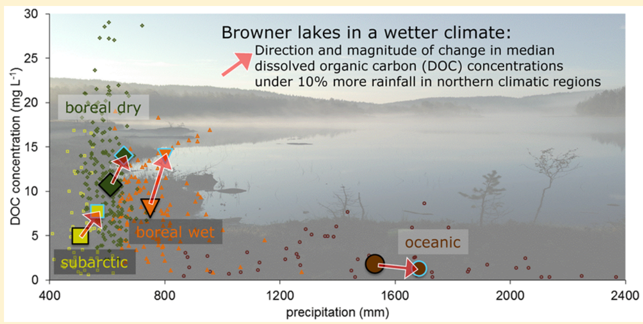
[∥]Finnish Environment Institute, P.O. Box 140, 00251 Helsinki, Finland

[⊥]Section for Aquatic Biology and Toxicology, Department of Biosciences, University of Oslo, 0316 Oslo, Norway

[#]Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, 901 83 Umeå, Sweden

Supporting Information

ABSTRACT: Browning of surface waters because of increasing terrestrial dissolved organic carbon (OC) concentrations is a concern for drinking water providers and can impact land carbon storage. We show that positive trends in OC in 474 streams, lakes, and rivers in boreal and subarctic ecosystems in Norway, Sweden, and Finland between 1990 and 2013 are surprisingly constant across climatic gradients and catchment sizes (median, +1.4% year^{-1}; interquartile range, +0.8−2.0% year^{-1}), implying that water bodies across the entire landscape are browning. The largest trends (median, +1.7% year^{-1}) were found in regions impacted by strong reductions in sulfur deposition, while subarctic regions showed the least browning (median, +0.8% year^{-1}). In dry regions, precipitation was a strong and positive driver of OC concentrations, declining in strength moving toward high rainfall sites. We estimate that a 10% increase in precipitation will increase mobilization of OC from soils to freshwaters by at least 30%, demonstrating the importance of climate wetting for the carbon cycle. We conclude that upon future increases in precipitation, current browning trends will continue across the entire aquatic continuum, requiring expensive adaptations in drinking water plants, increasing land to sea export of carbon, and impacting aquatic productivity and greenhouse gas emissions.



■ INTRODUCTION

Surface water browning from increasing terrestrial dissolved organic carbon (OC) concentrations[1] is a serious environmental concern. Browner surface waters may channel more greenhouse gases to the atmosphere,[2] lead to higher drinking water production costs,[3] and affect freshwater productivity through limiting light penetration[4] and creation of a more stable thermal stratification,[5] promoting anoxia and thereby limiting oxythermal habitats.[6] Increasing surface water OC concentrations in boreal and temperate regions in Fennoscandia (Norway, Sweden, and Finland), the United Kingdom, the northeastern United States, and southeastern Canada during recent decades have been extensively documented.[1] The majority of water bodies with positive OC trends appear to be in relatively small catchments,[1] while evidence of rising OC levels from larger water bodies is more ambiguous.[7] Changes in atmospheric chemistry, notably reduced sulfate deposition and variations in sea salt deposition, are a well-known driver for increased OC concentrations in boreal and temperate headwaters,[1,8,9] acting through chemically controlled organic matter solubility in catchment soils.[1,8−11] Discharge[12,13] and reduced processing related to shorter water residence times[14] may explain temporal and spatial OC variation.[9,15] Spatial and temporal variation in drivers makes predictions of future OC concentrations across a wide range of environmental gradients challenging. So far, an assessment of OC trends across water body types and catchment size is lacking.

Here, we assembled OC concentration data from 290 lakes and 184 running waters in Fennoscandia, ranging from headwater streams and lakes to major river basins, for the period from 1990 to 2013. The sites span wide gradients of sulfur deposition[16,17] and precipitation. We quantified trends and interannual variation of OC and use these to evaluate

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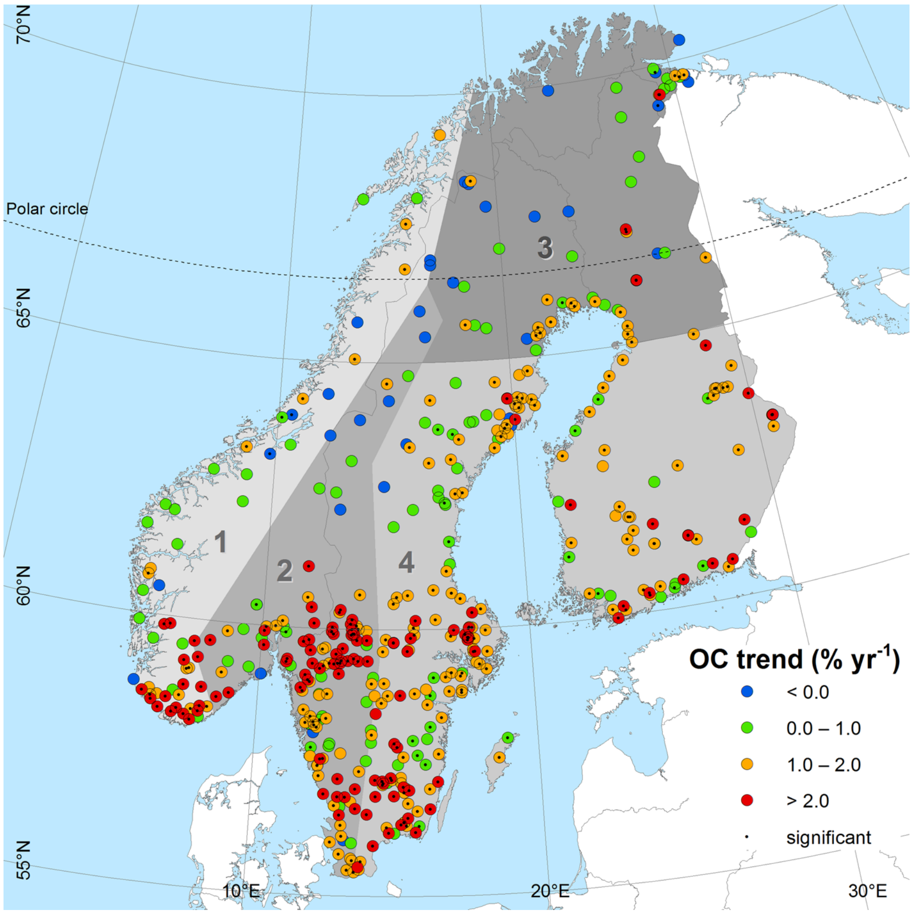

Figure 1. Organic carbon (OC) concentration trends (% year^{-1}) in 474 sites in Fennoscandia for the period from 1990 to 2013, divided across four regions (1, oceanic; 2, boreal wet; 3, subarctic, 4, boreal dry). Significance level of 0.05.

whether (i) OC concentration trends in water bodies depend on catchment size, water body type, sulfur deposition, and climate region and (ii) precipitation is a homogeneous driver of OC across climatic gradients.

MATERIALS AND METHODS

Water Chemistry. We collected OC concentration data for 474 surface waters sites (headwaters, streams, and rivers) with at least 15 years of data for the period from 1990 to 2013, in Norway (101 sites), Sweden (286 sites), and Finland (87 sites). OC was measured as total organic carbon (TOC), which in this region consists of 95 ± 5% of dissolved organic carbon (DOC).[18] Data for small headwater lakes and lower-order streams were taken from national acid deposition monitoring and inland water monitoring programs.[19−22] The catchments in these monitoring programs are selected for their lack of direct human disturbance such as settlements or agriculture. All monitoring programs follow well-established methods and analytical procedures.[7,16,19,21,23] River data were obtained from programs that monitor concentrations and loads of elements to coastal areas from large river basins.[21] Sampling frequency was, on average, four times per year but ranged from annually to quarterly, monthly, and weekly, usually with the highest sampling frequency for streams and rivers. Occasionally, the sampling frequency changed during the monitoring period. To avoid bias in trend estimates resulting from misrepresentation of seasonal variation, we included only data from seasons that were sampled throughout the monitoring period. Catchment size was available from national sources. Water chemistry records are publicly available at http://vannmiljo.miljodirektoratet.no (Norway), http://webstar.vatten.slu.se/db.html (Sweden), and https://wwwp2.ymparisto.fi/scripts/oiva.asp (Finland).

Precipitation. Annual precipitation records were available for 434 sites (four Norwegian and 36 Finnish sites missing). For Swedish and Finnish sites, precipitation data were obtained from the weather stations closest to the sampling location (Swedish Meteorological and Hydrological Institute and Finnish Meteorological Institute, respectively). For Norway, sampling coordinates were matched with precipitation grids of 0.5° × 0.5° available from the Climatic Research Unit (CRU, www.cru.uea.ac.uk/data) database.

Grouping of Sites. Sites were attributed to regions that were comparatively homogeneous with regard to sulfur (S) deposition and precipitation, with a requirement to include at least 10% of all sites. A Fennoscandian map of precipitation normals[24] was used to categorize sites in four precipitation classes. A map of wet deposition[17,25] was used to assign sites to three classes of sulfur deposition [i.e., low, <1 kg of S ha^{-1} year^{-1} (>65° latitude); high, 2−10 kg of S ha^{-1} year^{-1} (<62° latitude); and intermediate (62−65° latitude)]. The four regions were classified as subarctic (<700 mm year^{-1}, low deposition), boreal dry (700−1100 mm year^{-1}, intermediate to high deposition), boreal wet (1100−1400 mm year^{-1}, intermediate to high deposition), and oceanic (>1400 mm year^{-1}, intermediate to high deposition).

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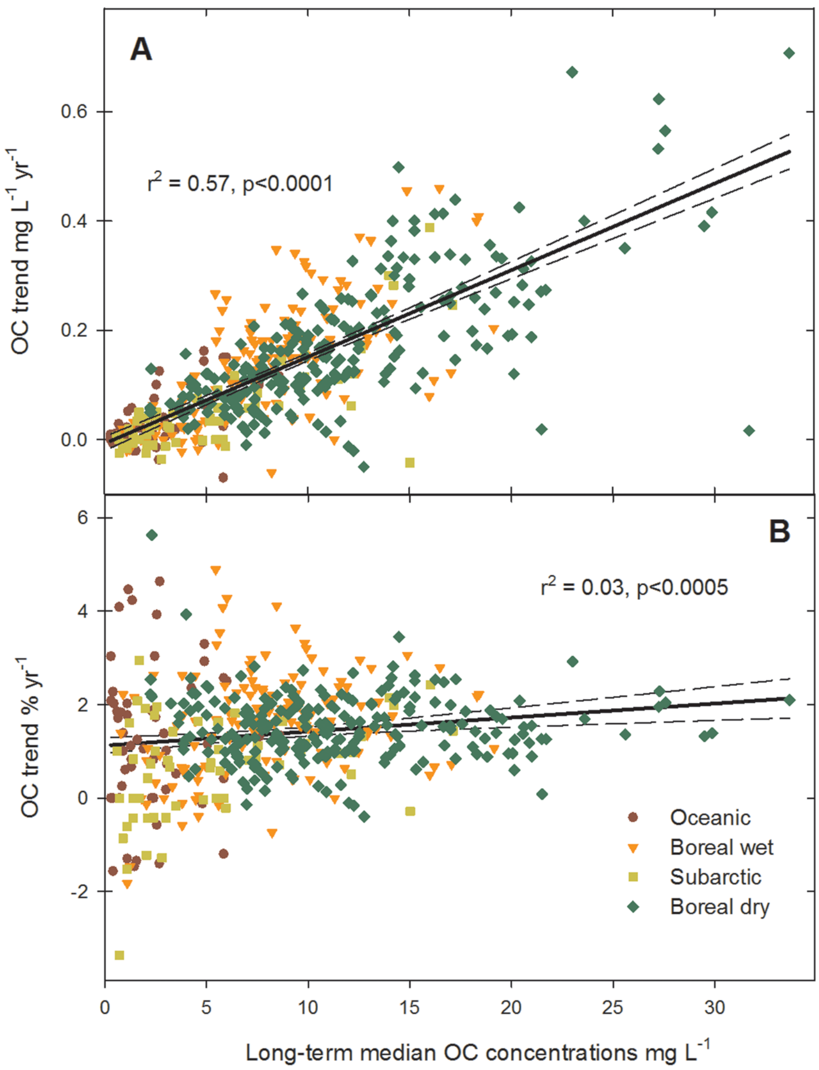

Figure 2. Relationships between trends of OC in 474 sites in Fennoscandia [(A) absolute and (B) relative] and long-term median OC concentrations, between 1990 and 2013. The color codes refer to oceanic, boreal wet, boreal dry, and subarctic Fennoscandia. Lines show linear relations, with 95% confidence intervals.

Calculations. The annual median OC concentration ({OC}) was calculated using the monthly or seasonal median {OC} in sites with multiple samples per year. All statistical tests were run in JMP, version 11, controlled for non-normal distributions by using median values and applying non-parametric tests such as the Wilcoxon and Mann−Kendall tests. For trend analyses, we used the Mann−Kendall test,[26] which gives a measure of significance of the long-term change of a variable ($p < 0.05$). We applied the Mann−Kendall trend test to yearly median {OC} and used the Theil−Sen estimator to estimate temporal trends in milligrams per liter per year (ΔOC). To transform absolute changes into relative changes (%ΔOC), in percent per year, we multiplied Theil−Sen slopes by 100 and divided by the long-term median {OC}.

RESULTS AND DISCUSSION

Boreal and subarctic surface water {OC} continues to increase significantly ($p < 0.05$) in the majority (67%) of sites across all of Fennoscandia, almost without exception (Figure 1 and Table S1). Even when a stricter criterion for significance ($p < 0.01$) is applied to lower the chance of false positives, 58% of the sites had significant positive trends. Such consistency across the landscape is a strong indication that global and regional, rather than local,[27] drivers are responsible for surface water browning. Water bodies with the highest {OC} had the highest trends, illustrated by the strong linear relationship between ΔOC and long-term median {OC} (Figure 2a; $r^2 = 0.57$; $p < 0.0001$). %ΔOC was almost insensitive to long-term median {OC} (Figure 2b; $r^2 = 0.03$; $p < 0.01$).

The median %ΔOC for Fennoscandia was 1.4% year^{-1} (interquartile range of +0.8−2.0% year^{-1}). The median %ΔOC and trend significance declined along a north to south gradient (Figure 1 and Table S1). The northward decline in %ΔOC is consistent with previously observed patterns in OC trends for the 1990s[1] and coincides with the strongest regional decreases in sulfur deposition that are found in southern Fennoscandia.[16,25] The currently assumed link between OC and sulfur deposition is a chemical control of organic matter solubility in catchment soils.[1,8−10] Since 1990, anthropogenic sulfur emissions in Europe have declined by 70%, and in the future, an additional 10% reduction is expected,[28] implying that the future impact of declining sulfur deposition on OC may be limited compared with past impacts. However, despite the relatively small change in sulfur

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deposition in the subarctic,[16] more than 40% of its sites had significant, positive OC trends (Table S1). Upward OC trends in the subarctic during the 1990s were significantly correlated with downward trends in sulfate,[1] but determining whether this driver or other mechanisms control OC trends after 2000 requires further analysis.

Catchment size (log-transformed, square kilometers) explained little variation in %ΔOC (r^2 = 0.02; p < 0.01). The negative relation between %ΔOC trend and catchment size existed for only lakes, not for running waters [r^2 = 0.03; p < 0.005 (Figure S1)]. While headwater OC is expected to be governed mostly by catchment processes [e.g., production, decomposition, (de)sorption, and lateral transport],[29] OC in water bodies with longer residence times is becoming increasingly impacted by in-lake and in-stream processing.[30,31] The decline of %ΔOC with lake catchment size but not with river catchment size is consistent with longer water residence times commonly found in lakes compared to those found in rivers.[31,32] Still, the low impact of catchment size on %ΔOC indicates a predominantly homogeneous rise in OC across the aquatic continuum in the hydrologically well-connected Fennoscandian landscape.

Variations in OC trend strength within and between climatic regions led us to explore responses of OC to rainfall. Along a precipitation gradient, we found strong relationships between the change in median annual {OC} and median annual precipitation, for groups of sites within defined intervals of mean annual precipitation (Figure 3). With the value expressed

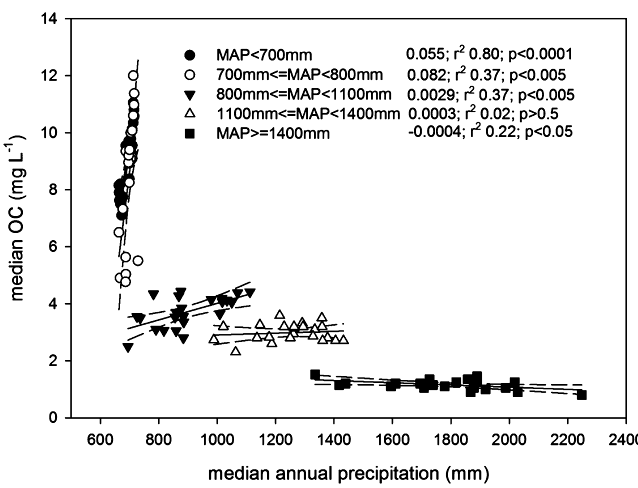

Figure 3. Linear relations between median of annual OC and the median of annual precipitation for groups of sites, grouped by long-term precipitation range (<700, 700−800, 800−1100, 1100−1400, and >1400 mm mean annual precipitation). Linear relations are plotted, with 95% confidence intervals. Medians calculated as the median of each year for all sites in a group. The legend shows the slope (mg of OC per liter per mm of precipitation), r^2, and significance levels.

as the ratio of the percent change in {OC} to a 10 mm change in precipitation, we found significant, highly positive ratios for the driest sites (+6% {OC}/10 mm, <700 mm interval; +8% {OC}/10 mm, 700−800 mm interval), significant and positive (+0.3% {OC}/10 mm, 800−1100 mm interval), significant and negative (−0.04% {OC}/10 mm, >1400 mm interval), and no significant relation (1100−1400 mm interval). This pattern suggests considerable mobilization of OC from increased precipitation at drier sites, declining toward wetter sites, until additional rainfall dilutes OC at the high end of the precipitation gradient. Similarly, large increases in the OC level in extremely wet years were found in boreal lakes in the northeastern United States.[33] Patterns of gradually declining OC mobilization and eventual dilution of OC with increasing levels of precipitation have also been shown by experimental wetting experiments.[34] Appreciating differential impacts of precipitation on {OC} along a climatic gradient leads to nuanced inferred negative effects of rainfall on {OC} from synoptic studies[35] and is an important step in improving regional predictions of future water quality.

Projections of future precipitation in Fennoscandia based on a large set of emission scenarios and global climate models agree on increases in precipitation but show a wide range of expected rates, from a 0.3−3% increase per decade[36] to a 5−30% increase in 2100.[37] This suggests that a 10% increase in precipitation in the course of 30 to 50 years is a reasonable expectation. Combining a 10% increase in annual precipitation with the relationships between {OC} and annual precipitation from Figure 3, we project substantial increases in {OC} across Fennoscandia, with the exception of oceanic regions (Figure 4).

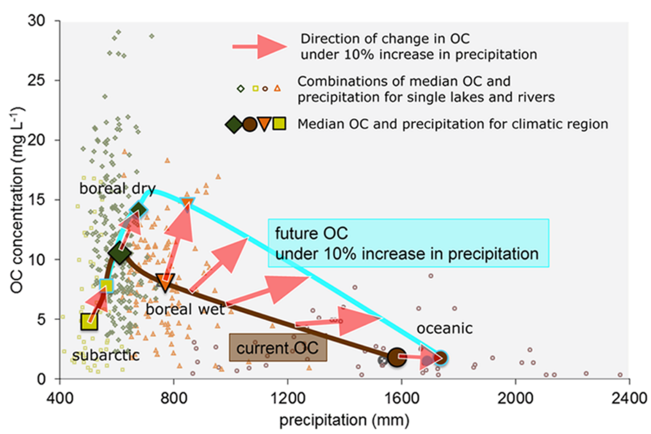

Figure 4. Projections of regional responses of OC concentrations to a wetter climate for climatic regions in Fennoscandia. The color coding refers to regions (oceanic, boreal wet, boreal dry, and subarctic). Small symbols show the median OC concentration and median annual precipitation for individual sites. Large symbols (marker lined in black) show the current median OC and precipitation in each region. Medium-sized symbols (markes lined in blue) show the change in medium OC for each region with a 10% increase in precipitation, estimated with the slopes derived in Figure 3 (chosen for the relevant precipitation range). The regionwise change in OC is indicated with gray arrows, while the assumed magnitude and direction of change in intermediate precipitation ranges are indicated with light brown arrows. For the sake of clarity, the current median OC is interpolated over the entire precipitation range with a brown line, and likewise for the future median OC but with a blue line.

In boreal dry Fennoscandia, we estimate an increase in median {OC} of +76%, twice as high as the increase in {OC} between 1990 and 2013 (+32%). In boreal wet and subarctic Fennoscandia, an additional 10% precipitation increases the median {OC} by +32% and +60%, respectively. In oceanic Fennoscandia, a 10% increase in precipitation results in a small dilution (−4%). For interpolated combinations of median {OC} and precipitation, we show possible trajectories of changes in {OC} given higher precipitation, assuming that OC mobilization declines with higher rainfall. All future {OC} projections remain within current ranges of precipitation and

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{OC} in Fennoscandia, suggesting that our results are plausible. However, the spatial variation in {OC} within each region is considerable and may increase further in a wetter climate.

Our projections indicate a 30−50% rise in {OC} in wetter climates. We interpret this primarily as an effect of hydrology, i.e., increased lateral flows through carbon-rich top soil layers[37,38] thereby increasing the rate of export of OC from catchment soils to freshwater bodies.[38,39] In addition, there are strong indications that a wetter climate may reduce rates of OC decay in freshwaters in boreal forests and tundra ecosystems by approximately 5−10%, as a result of less water retention and less time for aquatic cycling of OC.[31] Both mechanisms promote browning of surface waters and increase the rate of lateral transport[7] of terrestrially derived organic matter along the aquatic continuum.

The surface waters in our study are representative of a wide range of water bodies in northern Europe, including the United Kingdom and Russian Kola and Karelia,[40] and North America.[1,16,41] Thus, our findings suggest further browning in boreal and subarctic rivers and lakes in the Northern Hemisphere, given that dominating climate change projections are wetting rather than drying.[31] For Fennoscandia, we conclude that in a wetter climate, current browning trends will continue, except in oceanic regions currently experiencing precipitation above 1400 mm $year^{-1}$. On a regional scale, boreal Fennoscandia has the highest {OC} and will thus be impacted most.

Where drinking water utilities already experience extra costs for removal of higher {OC} during water treatment,[3,42] these cost increases will continue, possibly requiring new investments for higher capacity or novel treatment technologies. Also, elevated concentrations of mercury in fish exceeding limits for human consumption[43] are primarily found in humic waters and may increase, especially where browning results in changes in primary production[4,44] and fish growth.[45] Other possible impacts of continued browning are lower oxygen concentrations in lakes[6,46] and increased greenhouse gas emissions.[47,48] There is a pressing need for better integration of research about terrestrial and aquatic carbon, nutrient, and pollutant dynamics in carbon-rich boreal and subarctic ecosystems, encompassing process-based projections of surface water quality, element transport, and carbon sink functions in a future climate.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.6b00396.

Summary of OC trends grouped according to latitude, region, and catchment size (Table S1) and a plot of OC trends versus catchment size (Figure S1) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: heleen.de.wit@niva.no. Phone: +47 2218 5100. Fax: +47 2218 5200.

ORCID

Heleen A. de Wit: 0000-0001-5646-5390

Notes

The authors declare no competing financial interest.

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