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Lake Dissolved Organic Matter Quantity and Quality

Variability across Temporal and Spatial Scales

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Abstract

Müller, R. A. 2015. Lake Dissolved Organic Matter Quantity and Quality. Variability across Temporal and Spatial Scales. *Digital Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology* 1225. 37 pp. Uppsala: Acta Universitatis Upsaliensis. ISBN 978-91-554-9163-5.

Surface waters receive large amounts of dissolved organic matter (DOM) via runoff from land. The DOM is rich in organic carbon that serves as an energy source for the aquatic biota. During uptake of this energy, aquatic organisms mineralize organic carbon. The resulting inorganic carbon is partially released to the atmosphere as carbon dioxide and methane that are greenhouse gases, and which are of concern for the ongoing global warming. The rate at which organic carbon is mineralized depends strongly on DOM quantity and quality that vary with respect to both time and space. In this thesis, DOM quantity and quality were addressed using spectroscopic methods that build on the absorptive and fluorescent properties of chromophoric DOM (CDOM). New techniques to measure CDOM absorption and fluorescence were applied and further developed that allowed us to present novel CDOM variability patterns. Addressing the lake-rich Scandinavian landscape, strong focus was placed on water retention by lakes that tightly links to lake DOM quantity and quality.

An analysis of 24,742 lakes from seven large Swedish river systems indicated that the majority of lakes in Sweden exchange their water within one year. From headwaters to the Sea, summed lake volumes in the catchments of lakes were found to increase at rates comparable to discharge, which indicated effective water renewal along flow. A strong relationship between lake water retention and CDOM was apparent and further investigated based on samples from a lake district to a regional scale.

Results from *in situ* high-frequency monitoring of CDOM absorption in a eutrophic humic lake showed intra-annual variability patterns known from oligotrophic lake systems. The patterns for CDOM absorption contrasted results obtained for synchronously measured partial pressures of carbon dioxide that showed diurnal signals. Measurements of CDOM fluorescence and DOC concentrations indicated lake-internal DOM production. A comparison of these results with results from addressing 560 lakes distributed across Sweden, showed that a well-calibrated CDOM fluorescence measurement captures signals from lake-internal DOM production. I conclude that improved CDOM fluorescence measurements are promising to address lake-internally produced DOM.

Keywords: dissolved organic matter, organic carbon, CDOM, lakes

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To Uppsala

List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.

I Müller, R.A., Futter, M.N., Sobek, S., Nisell, J., Bishop, K. and Weyhenmeyer, G.A. (2013) Water renewal along the aquatic continuum offsets cumulative retention by lakes: implications for the character of organic carbon in boreal lakes. *Aquatic Sciences, 75*(4), 535-545, doi: 10.1007/s00027-013-0298-3.

II Kothawala, D.N., Stedmon, C.A., Müller, R.A., Weyhenmeyer, G.A., Köhler, S.J. and Tranvik, L.J. (2014) Controls of dissolved organic matter quality: evidence from a large-scale boreal lake survey. *Global Change Biology, 20*(4), 1101-1114, doi: 10.1111/gcb.12488.

III Müller, R.A., Kothawala, D.N., Podgrajsek, E., Sahlée, E., Koehler, B., Tranvik L.J. and Weyhenmeyer, G.A. (2014) Hourly, daily, and seasonal variability in the absorption spectra of chromophoric dissolved organic matter in a eutrophic, humic lake. *Journal of Geophysical Research: Biogeosciences, 119*(10), 1985-1998, doi:10.1002/2014JG002719.

IV Müller, R.A., Weyhenmeyer, G.A., Haaland, S. and Riise, G. Coherent color increases among 24 heterogenous lakes in a poorly-buffered lake district. *Manuscript*.

List of additional Papers

Outside this Thesis, the author has contributed to the following work.

- Weyhenmeyer, G.A., Kortelainen, P., Sobek, S., Müller, R.A. and Rantakari, M. (2012) Carbon dioxide in boreal surface waters: a comparison of lakes and streams. *Ecosystems, 15*(8), 1295-1307, doi:10.1007/s10021-012-9585-4.

- Weyhenmeyer, G.A., Müller, R.A., Norman, M. and Tranvik, L.J. Sensitivity of browning of boreal inland waters to future climate change. *Manuscript, in revision*.

Contents

Abbreviations

C	carbon
CDOM	chromophoric dissolved organic matter
C1-C6	fluorescence components
DOC	dissolved organic carbon
DOM	dissolved organic matter
DON	dissolved organic nitrogen
EEM	excitation-emission matrix
GHG	greenhouse gas
IC	inorganic carbon
N	nitrogen
P	phosphorus
pCO_2	partial pressure of carbon dioxide
TOC	total organic carbon
UV	ultraviolet
WRT	lake water retention time
%C1-%C6	relative abundance of the respective fluorescence components C1-C6
%Water	percentage of lake catchment area covered by water

Prologue

The results from my thesis primarily arise from field-based observations. Inherent to any field observation is the concern about scale, with respect to both time and space. We are bound to ask ourselves at which temporal and spatial scales variability predominantly occurs. For instance, we may enjoy swimming during summer, even at night, but it is far too cold during winter. We therefore can recognize a daily and a seasonal component that is characteristic for lake water temperatures. The latter exceeds the first in magnitude. Our observations are bound to spatial scales: a sampling site, a lake, a lake district, or broader regional and global scales. In summary, and for ecosystem properties that are accessible to our senses, we may to some extent trust our experience that grows so wonderfully free from math.

But what about ecosystem properties that are less accessible to our senses, and for which artificial sensors are merely about to be developed? Interactions between solutes and light that occur beyond the visible, gas and nutrient fluxes, are examples. Despite their fundamental role for lake ecosystem functioning, our senses are quite blind to their magnitude and variability. We are challenged to measure these ecosystem properties in representative ways, identify hotspots, and find ourselves confronted with large uncertainties. Facing these uncertainties, accessing variability patterns is a valuable task. Known variability patterns from the more directly perceptible ecosystem properties, like temperature, can serve as a useful reference. Also, it is here where the newly available large data that currently flood our lives help tremendously.

Reportedly, Benjamin Disraeli (British Prime Minister, 19^{th} century) said ‘Change is inevitable. Change is constant,’ which is analogous to philosophy taught by Confucius (孔子, chinese philosopher, 500 B.C.) and Heraclitus (greek philosopher, 500 B.C.). If there is some consistency in change, I can argue that an understanding of variability will lead to a more robust scientific theory.

Introduction

Carbocentric limnology

Freshwaters of the northern temperate and boreal region receive large amounts of organic matter from terrestrial soils via runoff (Thurman 1985). In boreal surface waters, the terrestrially derived (allochthonous) organic matter is primarily dissolved (DOM) and rich in carbon. The dissolved organic carbon (DOC) literally fuels the aquatic system with energy (Thurman 1985; Amon and Benner 1996). This energy is used by the aquatic biota, which had been recognized in the 1990's, and had been summarized by Wetzel (1995), cited by Steinberg (2003):

> 'Population fluxes are not representative of the material and energy fluxes of either the composite pelagic region or the lake ecosystem. Metabolism of particulate and especially dissolved organic detritus from many pelagic and non-pelagic autochthonous and from allochthonous sources dominates both material and energy fluxes.'
>
> (Wetzel 1995)

At the center of these material and energy fluxes stands the utilization of DOC by bacteria that re-introduces the dead organic matter into the food chain of the living (Tranvik 1992). The recognition of carbon as an energy source for the aquatic biota can be considered a paradigm shift within limnological sciences that led away from considering nitrogen (N) and phosphorus (P) as the two sole nutrients, to a concept of three prime nutrients that includes carbon (C). Lake ecosystem functioning could no longer be properly understood without at least a certain understanding of lake-internal DOC fluxes (Tranvik 1992; Findlay and Sinsabaugh 2003; Chapin et al. 2006; Prairie 2008; Guillemette and del Giorgio 2011).

The active role of freshwaters and lakes in global carbon cycling is an example. Biotic respiration and abiotic mineralization convert sizable amounts of DOC to inorganic carbon (IC). The resulting IC is partly emitted to the atmosphere as carbon dioxide and methane. In view of current climate warming projections, proper quantification and localization of the extent and relevance of this greenhouse gas (GHG) contribution from freshwaters serves as a central question to many carbon biogeochemistry studies (e.g. Melack et al. 2004; Cole et al. 2007; Battin et al. 2009; Tranvik et al. 2009; Raymond et al. 2013; Ran et al. 2014; Öquist et al. 2014). By 2013, the Intergovernmental

Panel on Climate Change (IPCC) first recognized estimates for GHG contributions arising from freshwaters that were forwarded to policymakers (IPCC 2013). The reported GHG emissions from inland waters sum to 1.0 Pg C yr^{-1}. These carbon emissions from inland waters to the atmosphere are comparable to the carbon load transported from land via freshwaters to the oceans, and exceeds the estimated carbon burial in lakes by a factor of five (IPCC 2013).

A new research field had developed that was aptly coined 'carbocentric limnology' by Jon Cole (Prairie 2008). In short, a carbocentric limnology addresses lake carbon cycling, the carbon balance, and its role for lake ecosystem functioning as well as water quality related issues. In this context, interactions between chromophoric dissolved organic matter (CDOM) and light play a central role (Thurman 1985; Kirk 1994).

CDOM absorption and fluorescence

Light absorption by CDOM affects the distribution of heat in the water column (Fee et al. 1996; Snucins and Gunn 2000), limits the depth of the photic zone that is essential for primary production (Kirk 1994), and limits the penetration of ultraviolet (UV) radiation that can harm aquatic organisms (Morris et al. 1995; Hargreaves 2003). Moreover, the color of water is affected by CDOM absorption in the visible range (390 to 700 nm) of the electromagnetic spectrum. As an example, recent increases in DOC concentrations have been associated with increased color (e.g. Hongve et al. 2004; Monteith et al. 2007), which is a central concern for lakes and reservoirs used for drinking water purposes (Siddiqui et al. 1997; Wilhelm 2009). For water quality monitoring, the absorptive and fluorescent properties of CDOM allow to apply spectroscopic methods to assess DOM quantity and quality aspects:

In terms of DOM quantity, measurements of CDOM absorption and fluorescence are used as a surrogate for DOC concentrations (e.g. Meili 1992; Green and Blough 1994; Kirk 1994). In more recent years, this spectroscopic approach allowed for continuous *in situ* high-frequency time series of DOC concentrations in flowing waters (e.g. Saraceno et al. 2009; Jeong et al. 2012; Sandford et al. 2013). This *in situ* spectroscopic approach is promising as it is cost-effective and relatively robust.

In terms of DOM quality, measurements of CDOM absorption and fluorescence are used to describe DOM composition. For this purpose, CDOM absorption and fluorescence signals occurring at specific wavelengths and wavebands along the UV and visible region, are being addressed and studied (e.g. Kirk 1994; McKnight et al. 2001; Cory et al. 2010).

Finally, measurements of CDOM absorption offer an objective standard for measuring water color (Kirk 1994). The measurements are objective in the sense that they are quantitative and reproducible. Cuthbert and Del Giorgio (1992) note that alternative, traditional methods to measure water color, like the Hazen method (DIN EN ISO 6271), require a subjective visual comparison with a standard solution that may be perceived differently by different observers, and in different light environments. This subjective source of human error is of little concern for CDOM absorption and fluorescence spectrometry that is nowadays applied worldwide.

For lake water bodies, different CDOM absorption and fluorescence properties are characteristic of DOM from terrestrial (allochthonous) and lake-internal (autochthonous) sources. In short, the terrestrial DOM is usually rich in fulvic and humic acids that are degradation products from plant structural materials, such as cellulose and lignin. Lake-internally produced DOM is rich in proteins and is frequently found to be less absorptive of light than its terrestrial counterpart (Thurman 1985; Steinberg 2003). In lakes, DOM from different sources mix and are subject to further lake-internal production and degradation. These lake-internal processes induce CDOM variability that expresses at specific temporal (e.g. daily, monthly) and spatial (e.g. point, lake) scales. For eutrophic systems, it is suggested that relevant CDOM variability occurs at short (hourly, daily) temporal scales, and during productive time periods (e.g. Spencer et al. 2007). Addressing short-term temporal scales demands *in situ* monitoring techniques that are merely about to be developed (Saraceno et al. 2009; Coloso et al. 2011; Liu et al. 2013). As a consequence, the *in situ* short-term variability in lake CDOM absorption and fluorescence is as of yet largely unknown. Finally, the lake-internal DOM mixing, production, and degradation is compounded by lake water renewal (e.g. Schindler 1990; Meili 1992).

Lake water retention and flow

Along surface water flow paths, and along lake chains, DOM quantity and quality are affected by retention in lakes (Soranno et al. 1999; Larson et al. 2007; Sadro et al. 2012; Weyhenmeyer et al. 2012). For example, Spencer et al. (2012) discuss that the presence of lakes in the catchment of large rivers weakens the relationship between CDOM and DOC concentrations that challenges the application of spectroscopic methods to estimate DOM quantity. Goodman et al. (2011) demonstrate that the presence of lakes upstream can buffer variability in DOC concentrations that gives a first cause for a weak relationship between CDOM and DOC concentrations, since correlative analyses perform best along strong DOC concentration gradients. Conversely, tributaries can effectively renew surface waters along flow (e.g. Alexander et al. 2007). Also, the local geology and the groundwater influence can vary strongly along a gradient of flow (Kratz et al. 1997; Magnuson et al. 2006). The positioning of lakes in the landscape, along with their relative volumes, and lake:catchment sizes are crucial for the accumulation of surface water retention that can be demonstrated applying simple numerical examples (Figure 1). For large hydrological networks, however, the accumulated retention by lakes and its influence on CDOM absorption and fluorescence is largely unknown.

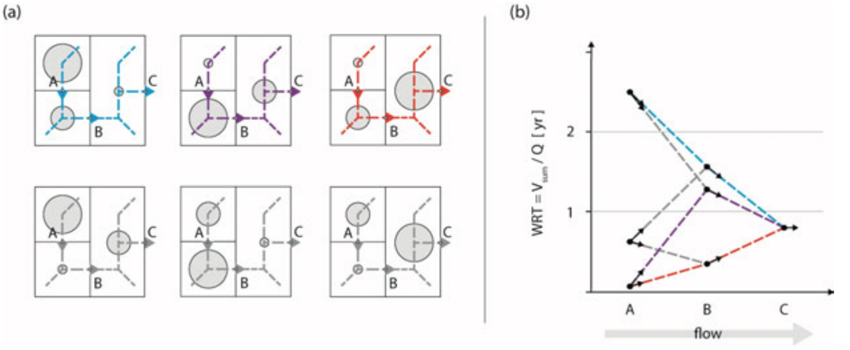

Figure 1. A theoretical drainage network with three lakes of different volumes, 0.1, 1.0, and 4.0 Mm^3 (grey circles). The lakes are placed in three localities within the landscape that can be permuted, which leads to a total of six possible lake landscape arrangements. The three sub-catchments of 4, 8 and 16 km^2 are shown in boxes, with their outflows denoted as A, B, and C, respectively (a). For simplicity, we assume runoff to be homogenous with 300 mm yr^{-1}. Discharge (Q) accumulates along flow that is via A (Q_A=0.05 m^3s^{-1}), to B (Q_B=0.10 m^3s^{-1}), and towards C (Q_C=0.20 m^3s^{-1}). Lake volumes accumulate along the lake chains. We calculated the summed lake volume (V_{sum}) to discharge (Q) ratio to indicate lake water retention (WRT) at a landscape scale for each of the six permutations (b). For both figure parts, three exemplary lake chains and their WRT are highlighted in colors (blue, purple, and red).

Aim of the Thesis

The overall aim of my doctoral thesis was to improve the understanding of DOM quantity and quality aspects for Swedish boreal lakes. The following main research question lies at the center of my thesis:

- Which processes control dissolved organic matter quantity and quality at different temporal and spatial scales?

The following more detailed research topics were raised and addressed:

- Lake water retention and its effects on dissolved organic matter
  Do DOM quantity and quality change from headwaters towards the Sea? (Paper I)
- Catchment control of dissolved organic matter
  Which water chemical and catchment variables control DOM quantity and quality at a lake-district to regional scale? (Paper II and IV)
- High-frequency monitoring of dissolved organic matter
  To which extent do lake-internal processes induce hourly, daily, and seasonal patterns in CDOM absorption?(Paper III)

Methods

Study sites

Spatial variability in DOM quantity and quality were addressed at a regional scale (Paper I and II). For this purpose, geomorphological and hydrological data from 24,742 lakes (Paper I), and 560 lakes (Paper II) were used that were available from the Swedish Meteorological and Hydrological Institute (SMHI). Water chemical data from 1,559 lakes (Paper I), and 560 lakes (Paper II) were used that were available from the Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences (SLU).

Temporal variability in DOM quantity and quality were addressed at a lake-district scale in Østmarke that is located near Oslo, Norway (Paper IV). The lake district is 135.3 km^2 in size and holds numerous lakes, of which 24 are sampled regularly by the Norwegian University of Life Sciences.

Temporal variability in DOM quantity and quality at hourly, daily, and seasonal time scales was addressed in a single lake system. For this purpose, an intensive monitoring program was conducted at lake Tämnaren (60°10′N, 17°20′E), a eutrophic, humic lake of temperate Sweden (Paper III). The study was supported with observations from 23 other local lakes within a similar climatic zone that were sampled by the SLU, during the same time period.

A schematic overview of the four individual studies comprising my thesis, and their relation to scales of time and space, is given in Figure 2.

Geomorphological and hydrological analyses

General water exchange characteristics of lakes in the Swedish landscape (Paper I-II), their catchment size, landcover, and landscape position, were investigated by applying spatial analyses that were run using geographic information systems (GIS). Lake water retention time (WRT) was estimated as the lake volume (m^3), divided by the long-term (1961-1990) mean discharge (m^{3}s^{-1}) measured at the lake outflow. Lake volume estimates were available from the Swedish Meteorological and Hydrological Institute (SMHI). Where volume estimates were missing, an empirical lake volume estimator was applied following methods presented by Sobek et al. (2011). Comparable methods were applied in Paper IV, where data was available from Norwegian authorities.

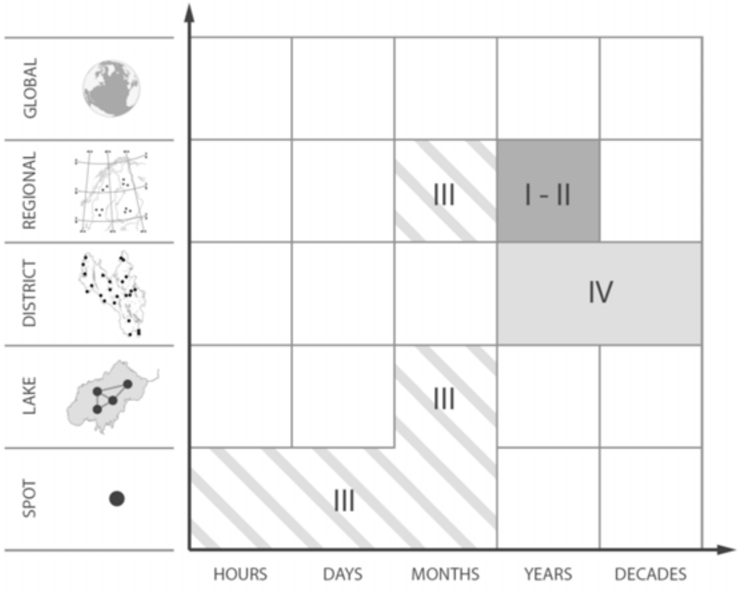

Figure 2. Schematic overview of the temporal (horizontal axis) and spatial (vertical axis) scales for which DOM quantity and quality aspects were addressed in the individual studies comprising this thesis (marked). Regional lake monitoring (Paper I-II), at a single lake, Tämnaren (Paper III), and in the Østmarka lake district (Paper IV).

Regional lake water sampling and analyses

Regional lake water sampling and analyses were conducted by the SLU (Paper I, II and III) and the Norwegian University of Life Sciences (Paper IV). Lake water chemical analyses were performed in accredited laboratories that encompass a wide variety of water chemical variables: base cations, anions, nutrients, including carbon contents, metals, including aluminium, iron and magnesium. In Paper I and IV, organic carbon contents were quantified on unfiltered samples as total organic carbon (TOC) concentrations and used as a proxy for DOC concentrations, which is an accepted method for boreal surface waters (e.g. Hongve et al. 2004; von Wachenfeldt and Tranvik 2008). Other variables used are specified in more detail in the individual Papers I-IV. To characterize CDOM fluorescence, a total of 560 samples from standard SLU lake monitoring conducted in the years 2009 and 2010, were analyzed locally at the Department of Ecology and Genetics/Limnology, Uppsala University (Paper II). Excitation-emission matrices (EEMs) and absorption spectra were measured applying CDOM fluorescence and absorbance spectrophotometry, respectively.

High-frequency monitoring at lake Tämnaren

We deployed and maintained a continuous monitoring *in situ* spectrophotometer probe from 6 May to 19 October, 2011, at a meteorological monitoring station located in the central part of Tämnaren. The *in situ* spectrophotometer probe measured beam attenuation at wavelengths spanning from 240 to 735 nm, at 2.5 nm intervals and across an optical path length of 0.035 m. I developed and applied a correction for particle interferences to estimate CDOM absorption from the beam attenuation measured. A collaboration with meteorological monitoring, run by the Department of Earth Sciences, Uppsala University, allowed for comparisons with physical time series, such as solar radiation, temperature, wind, and partial pressures of carbon dioxide (pCO_2) in water and air. Continuous *in situ* monitoring was validated with intense manual sampling. Three sites were sampled on 21 occasions for organic and inorganic carbon levels, plus optical analyses to address DOM quantity and quality, among other measures. Laboratory analyses were conducted at the Department of Ecology and Genetics/Limnology, Uppsala University.

Statistical analyses

Standard statistical analyses were applied to test the significance of results. Relevant concerns about the assumptions made when applying standard methods were respected; in specific, non-normality and heteroscedasticity (subgroups with unequal variability) in the data were considered (see e.g. Quinn and Keough, 2002). Where necessary, alternative statistical methods, e.g. nonparametrical methods, were applied in order to test significance. To rank predictors of the variability observed in DOM quantity and quality, multivariate statistical techniques were used. As an example, principle component (PCA) and partial least squares analysis (PLS) were applied to rank chemical, physical, and geographical predictors (Paper II and IV). Parallel factor analysis (PARAFAC) was applied to characterize CDOM fluorescence by identifying underlying fluorophore regions that vary independently of each other (Paper II). Generalized linear regression models were applied to correct *in situ* measurements on unfiltered lake water for particle interferences (Paper III). Mann-Kendall tests were applied to test the significance for long-term temporal trends (Paper IV).

Results and Discussion

Lake water retention and its effects on dissolved organic matter

(Paper I and II)

The Swedish National Lake Inventory counts an approximate 95,700 lakes larger 0.01 km^2 within the geographic extent of Sweden. Analysis of 24,742 (25.9%) lakes from seven large drainage networks (Lagan, Mälaren, Dalälven, Ljungan, Ångermanälven, Umeälven, and Kalixälven) in Sweden showed that the majority of lakes exchange their water within one year (Figure 3). In boreal Scandinavia, lake water renewal is driven by intense runoff events that include the yearly spring runoff due to snowmelt, but also strong rainfalls during summer and early autumn (Lindström and Bergström 2004; Ågren et al. 2010). Long-term (1961-1990) mean discharge at lake outlets accumulates effectively from headwater lakes towards lakes at low landscape positions. We found that the summed lake volumes in the catchments of lakes increased at a rate comparable to the discharge, which indicates effective water renewal along flow. Our findings are in accordance with earlier studies that report a weak relationship between water retention and catchment size for large drainage networks (Tokunaga 2003; Alexander et al. 2007). In addition, water may be renewed even more effectively than demonstrated by our results. An example is the groundwater influence that can be expected to lower DOM levels via dilution (groundwater exfiltration) and removal (groundwater infiltration).

Applying spectroscopic methods, we report high losses in CDOM absorption for lakes with WRTs below one year. Strong initial CDOM losses are frequently reported to accompany runoff from land (e.g. Meili 1992). It has been demonstrated that terrestrial DOM is destabilized as it is released from soils to freshwaters, and therefore becomes subject to degradation (Kalbitz et al. 2005; Kogel-Knabner et al. 2008). In agreement with earlier studies (e.g. Weyhenmeyer et al. 2012), we show a preferential removal of CDOM over DOC along the WRT gradient. Solar irradiance leads to direct photo-oxidation of CDOM that explains a selective removal of CDOM over DOC (Vähätalo and Wetzel 2004; Vähätalo 2010). In accordance, the terrestrial DOM becomes exposed to light as soon it emerges to surface waters. Conversely, solar irradiance drives photosynthesis and therein lake-internal DOM production. Consequently, sunlight alters DOM quantity and quality both via degradation and production. However, assessing the net-effect of sunlight on the lake organic carbon pool is challenging (Bertilsson and Tranvik 1998; Tranvik and Bertilsson 2001).

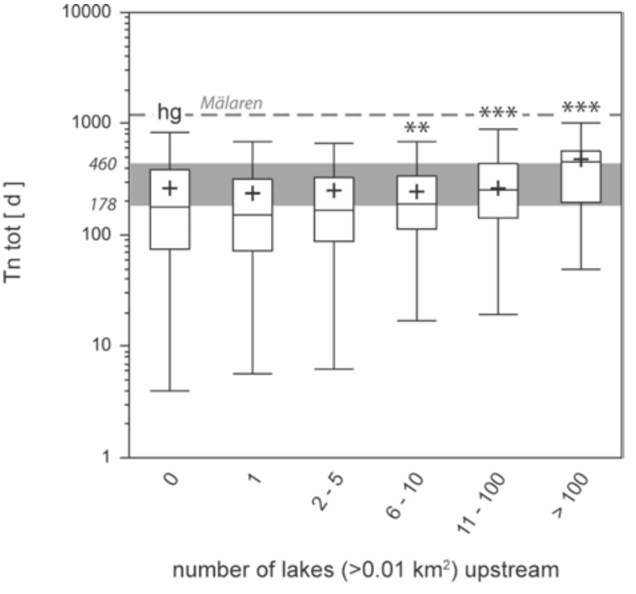

Figure 3. Lake volume (V_{sum}) in the catchment of each of 24,742 lakes, divided by the long-term (1961-1990) mean discharge (Q) at the lake outflow, as lake water retention time (WRT, here denoted $T_{n\ tot}$) and relative to landscape positioning, as indicated by the number of lakes upstream (>0.01 km^2). Box and whisker plots indicate the distribution (median, 50% and 90% quantile ranges) for WRT estimates in each group. The increase in median WRT from the headwater group towards the most downstream group is marked (dark-grey). Asterisks indicate significant differences (* $p<0.05$, ** $p<0.001$, *** $p<0.0001$) for an increase in the median WRT when compared to the headwater group (hg). Crosses (+) mark median WRT for lakes sampled. This figure has been reproduced from Paper I, with permission from the publisher. I here marked a WRT of 3 yr that is typical for the third largest lake in Sweden, Mälaren, located near Stockholm (dashed horizontal line).

A preferential removal of CDOM over DOC was not expressed from headwater lakes towards lakes at low landscape position, as CDOM and DOC decreased in parallel (Paper I). We argue that this pattern is supported by dilution that increases along flow, which can result from an increased groundwater influence towards low landscape positions (reviewed in Magnusson et al. 2006). The increase in WRT from headwater lakes (median=178 d) towards low landscape positions (median=460 d) was rather modest (Figure 3). We highlight that long WRT are reached in large individual systems. In these systems the time is given for lake-internal processes that can alter DOM quality substantially, as was demonstrated for Mälaren (1,090 km^2, WRT ca. 3 yr) in Köhler et al. (2013).

By applying CDOM fluorescence techniques, we investigated the role of water renewal for DOM quantity and quality aspects that reach beyond conventional measurements of CDOM absorption and DOC concentration. We found that both the WRT, and the percentage of the lake catchment area covered by water (%Water), strongly relate to the relative abundance of two key and contrasting fluorescence components (C3 and C6) in lake waters (Figure 4). Component C3 indicates a terrestrial, humic-like signal, which is lost most rapidly from water bodies and therewith shows similar patterns as CDOM absorption (Figure 4a, c, e, and g). The additional information gained from CDOM fluorescence in terms of lake water renewal mainly relates to the increasing proportion of protein-like component C6 with WRT, or %Water. C6 indicates lake-internal DOM production and processing. Examples are DOM production by algae and macrophytes that are highly relevant energy sources for the microbial community, and as such, for lake metabolism in general (e.g. Guillemette and del Giorgio 2011). We show that the relative contribution of the protein-like signal %C6 increases significantly with WRT and %Water (Figure 2b, 2d). Moreover, the relative contribution of C6 is high (>10%) in low DOC systems and becomes more and more suppressed towards humic-like, high DOC systems (Figure 4f).

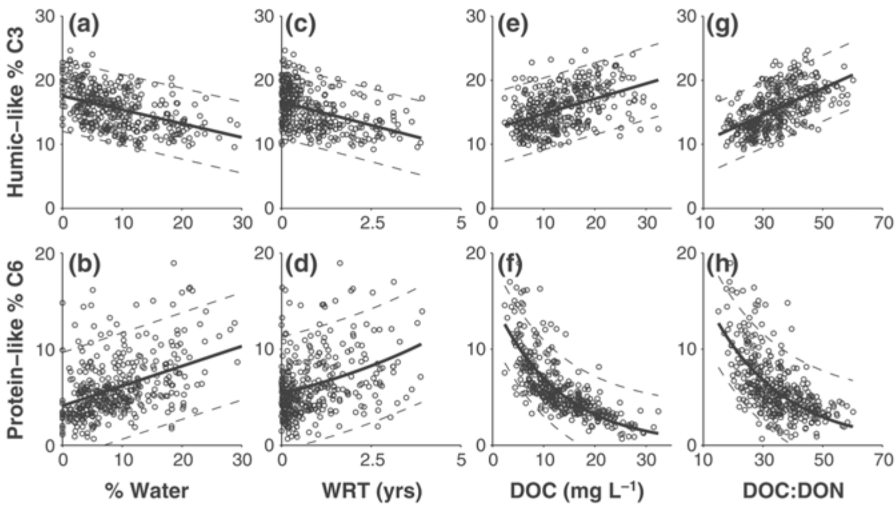

Figure 4. Relationship between the relative abundance of the fluorescence components C3 (as %C3) and C6 (as %C6), and the percentage of water in the surrounding catchment (%Water) (a, b), lake water retention time (WRT) (c, d), dissolved organic carbon (DOC) (e, f), and DOC to dissolved organic nitrogen (DOC:DON) (g, h). This figure has been reproduced from Paper II, with permission from the publisher.

In summary, Paper I and II highlight that lake-internal effects on DOM quantity and quality are offset by water renewal that is effective in the boreal region and facilitates the delivery of CDOM rich, terrestrial organic matter from catchment soils. In addition, we note that lake-internally produced protein-like CDOM fluorescence (C6) is suppressed in humic waters with high DOC concentrations (Figure 4f).

High-frequency monitoring of dissolved organic matter

(Paper III)

Events of lake-internal production can lead to changes in DOM quantity and quality that express at relatively short time scales (weekly, daily, hourly). To which extent spectroscopic methods are capable of tracking lake-internal DOM production, and aid in monitoring lake-internal carbon cycling, remains largely unknown (Coloso et al. 2011). Arguably, lake-internal production could induce diurnal CDOM signals during highly productive time periods (Spencer et al. 2007). Certainly, temporal responses of such form are well-known from dissolved oxygen and pCO_2 time series. In accordance with the theory, we report regular diurnal patterns for pCO_2 (water and air) that indicate photosynthetic production (Paper III). A regular diurnal signal for CDOM absorption was, however, not observed. Potentially, CDOM absorption signals are quenched in the humic lake Tämnaren, by stronger effects that associate with the CDOM delivery from land, and its subsequent degradation in the lake. We report continuous CDOM degradation over the summer months which was accompanied by changes in the shape of the absorption spectra that indicates a radiation induced CDOM loss. We support this argument with results from photobleaching rate modeling. For the first time, we were able to show that a continuous summer CDOM loss can exceed short-term (hourly to daily) variability by an order of magnitude, and over a full season.

Applying Fourier transform analysis, a method commonly used by climatologists, we were able to identify the scales (hourly, daily, monthly) at which temporal variability occurred (Figure 5). Multiple CDOM absorption metrics showed a nearly identical result; a decreasing temporal variability towards shorter temporal scales (Figure 5b). The Fourier transform analysis was especially valuable for comparing the CDOM absorption time series to known and unknown variability patterns in physical and meteorological time series. A daily to weekly signal was apparent for the light attenuation, which is the raw signal measured by the *in situ* spectrophotometer probe (Figure 5a). A daily to weekly signal was also apparent for wind (Figure 5d). For the shallow lake Tämnaren, we suggest that these daily to weekly patterns resulted from wind induced sediment resuspension that affects turbidity. These interactions with the sediment carbon pool did, however, not induce strong changes in CDOM absorption, which is likely due to the frequency of the wind-induced water column mixing that does not allow for anoxia, nor for a vertical CDOM gradient to develop (e.g. Downing et al. 2008).

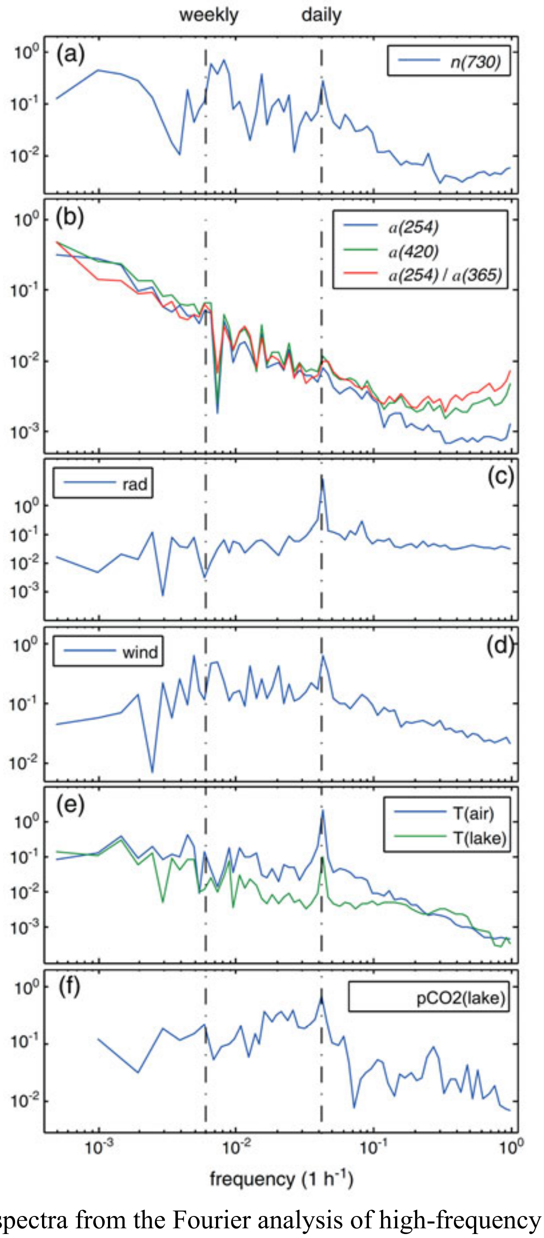

Figure 5. Power spectra from the Fourier analysis of high-frequency data, for beam attenuation from particle interferences (a), absorption coefficients as measures of CDOM (b), radiation (c), wind (d), lake water and air temperatures (e), and partial pressures of carbon dioxide in the lake water (f). The daily and weekly signal is indicated (labels on top, with dotted vertical lines). The data is from lake Tämnaren and was measured during the period of 6 May to 19 October, 2011. This figure has been reproduced from Paper III, with permission from the publisher.

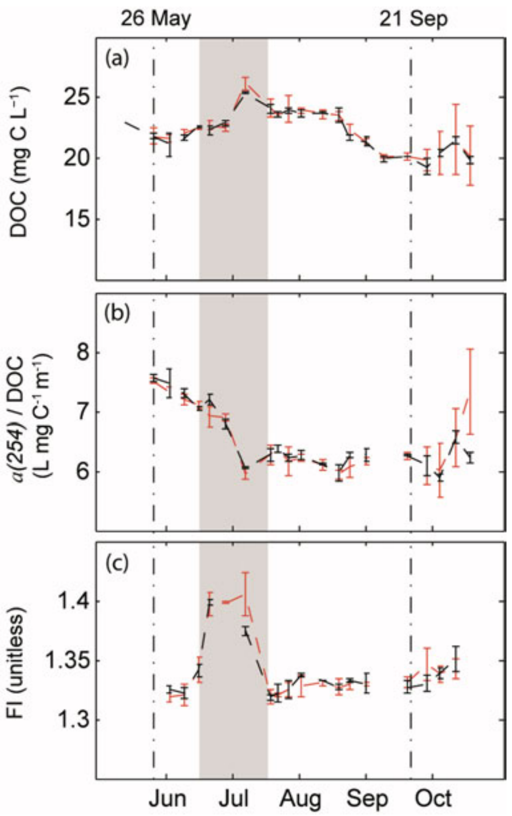

Figure 6. Summary from 21 sampling occasions for DOC concentrations (a), specific UV absorption as CDOM absorption, *a*(254), per unit DOC (b), and the fluorescence index (FI) (c). All three figure parts show medians and standard deviations for triplicates taken by the spectrophotometer at the float (black) and for three samples taken across a transect from the shore towards the float (red). The data are from lake Tämnaren and were measured during the period of 6 May to 19 October, 2011. This figure has been reproduced from Paper III, with permission from the publisher. We show three of six figure parts and highlight a productive time period from mid-June to mid-July (grey shaded).

Results from discrete sampling confirm strong differences for CDOM absorption and fluorescence signals. During a productive time period (Figure 6, in grey), the DOC concentration increased while absorption by CDOM, and the CDOM:DOC ratio, decreased. Conversely, the fluorescence index (FI) increased, suggesting more DOM from microbial and algal sources. The FI is a CDOM fluorescence metric designed to indicate lake-internal DOM production (McKnight et al. 2001; Cory et al. 2010). Spatial variability, as variability across sampling sites, was very low (<10%) during summer and increased strongly as autumn leaf fall initiated during late September 2011 (Figure 6, in red).

Coherence among lakes and dissolved organic matter

Paper IV

Addressing a lake district in southern Norway, and for a decadal (1983-2012) time scale, we investigated the inter-annual coherence among lakes for the DOM related water chemical variables TOC, iron (Fe), and color, along with 14 additional water chemical variables. We defined coherence as a synchronous response of multiple lakes to a defined driver (Magnusson et al. 2006), with the response measured at an inter-annual time scale. Coherence among lakes was used synonymously with synchronicity. At the lake-district scale, differences in regional drivers for color that include growing season length, temperature, precipitation amounts, and the atmospheric deposition chemistry, can be considered negligible. Still, multiple lakes with contrasting lake morphological and catchment characteristics can be compared. This makes a lake-district scale especially well-suited for addressing the catchment influence on lake DOM.

For the Norwegian lake district that is subject to significant long-term increases in color (Hongve et al. 2004), we show that the inter-annual coherence among 24 lakes differed strongly for different water chemical variables. Sampling was conducted during autumn, when the catchment influence is often stronger than during the preceding summer months. The catchment influence was expected to induce variability that differs between systems, and generally tends to lower coherence among lakes. This is especially so for the variables TOC, Fe, and color, which are subject to a strong catchment influence. We report a moderate coherence for color (Figure 7), which is comparable to results by Pace and Cole (2002), who addressed coherence in color for a suite of 20 lakes in northern Michigan, USA. For the 24 lakes sampled in the Norwegian lake district, we found that the count of lakes with significant long-term (1983-2012) temporal trends related strongly to the coherence reported for the individual water chemical variables (Figure 7). Based on this result, I argue that a low inter-annual coherence among lakes can affect the frequency with which underlying long-term temporal changes are detected. In terms of water chemical variables that describe DOM quantity and quality, we show that the inter-annual coherence among lakes is higher for color than it is for TOC and iron, suggesting that long-term temporal changes in color are most readily disclosed (Figure 7).

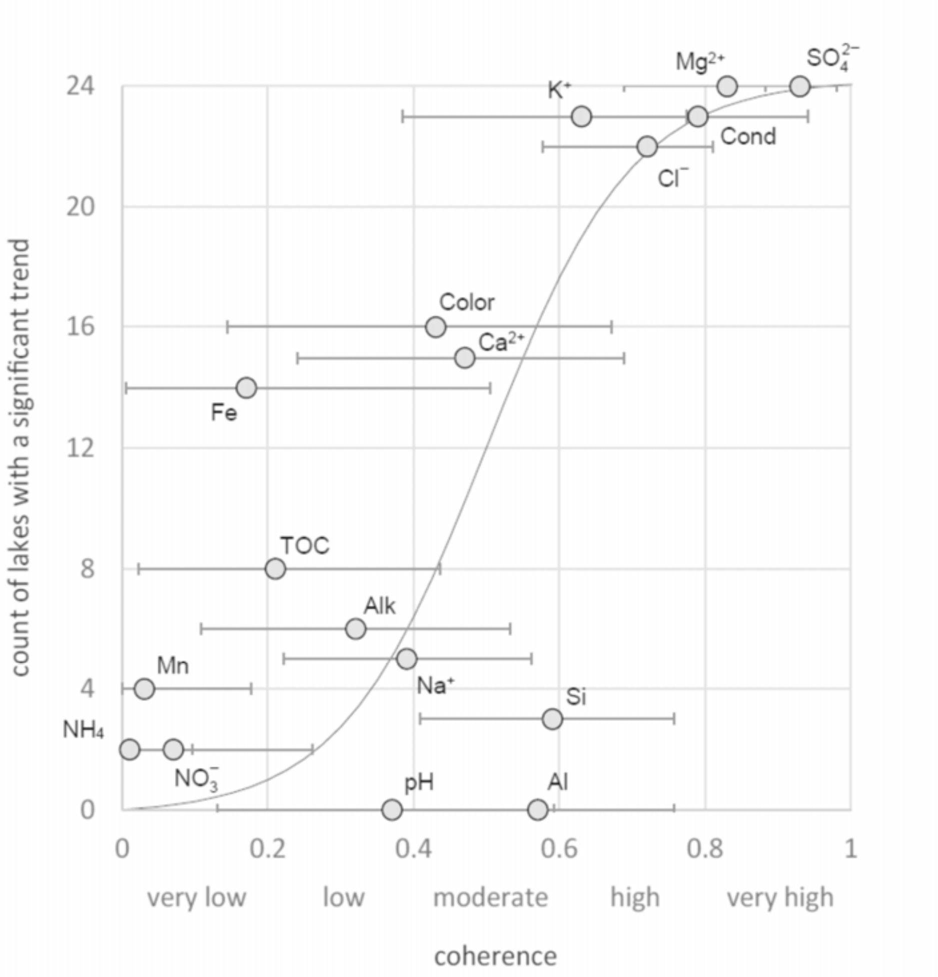

Figure 7. Comparison of inter-annual coherence among lakes, with the count of lakes for which a significant temporal trend was reported. The count of lakes is indifferent to the direction (positive, negative) of the trend, as long as significance at $P<0.05$ was reached. Whiskers indicate the 50% quantile ranges for R^2s from coherence analyses. The results are presented for each water chemical variable measured (N=17). For the Norwegian lake district addressed, the TOC concentrations can be used as a proxy for DOC concentrations (Hongve et al. 2004).The s-curve (black line) is purely conceptual (not based on actual data).

Conclusions and Perspectives

Paper I demonstrated that the vast majority of lakes in the Swedish landscape are regularly flushed with reactive terrestrial DOM. Based on data from numerous (>1,000) lake systems, we show decreasing CDOM absorption for increasing lake WRTs. Water renewal was effective along flow, even along well-connected drainage systems that cut across strong environmental gradients. Our results support earlier findings that show a weak relationship between lake landscape positions and WRT. Effective water renewal along flow may allow local climate and landscape characteristics to express effectively in individual lake systems.

I give an example of decreasing CDOM absorption (Figure 6b) for a period where CDOM fluorescence (Figure 6c) and DOC concentrations (Figure 6a) increased, which indicates lake-internal DOM production. Based on these results, I argue that a lake-internal DOM production is poorly documented by measurements of CDOM absorption. Also, and in accordance with other studies, we show that lake-internal processes lead to a disconnection between CDOM absorption and DOC concentrations. In contrast, a protein-type CDOM fluorescence metric is sensitive to the lake-internal DOM production (Figure 6c). In addition, in Paper II, we successfully identified a protein-type signal component in the bulk CDOM fluorescence matrix, applying multivariate signal deconvolution (Figure 4). From Paper II and III, I conclude that a well-calibrated CDOM fluorescence metric offers a potential means to track events of lake-internal DOM production.

For future studies that target the delivery of terrestrial DOM to lake systems, I suggest to use the humic-type CDOM signal that can be quantified applying robust CDOM absorption techniques. Regarding *in situ* CDOM absorption probes, the measurement path-length, clean optical lenses (biofoul), and a correction for particle interferences are critical. Addressing CDOM absorption, a filtration step may be avoidable even in turbid lake systems (Paper III).

For future studies that target a lake-internal DOM production, I suggest to address the protein-type CDOM fluorescence signal. For the purpose of tracking a protein-type DOM component, CDOM fluorescence probes are needed that are calibrated respectively, and are suitable for measuring lake-internally produced DOM *in situ*, and at a high-frequency interval.

For future studies that address changes in DOM quantity and quality at decadal time scales, I suggest to account for differences in the inter-annual coherence of individual variables measured (e.g. TOC, Fe, color), as such affects the statistical significance, and there with the detection and interpretation of long-term temporal trends (Figure 7).

Summary in Swedish

Mängd och sammansättning av löst organiskt material i sjöar

(variabilitet i tid och rum)

Sjöar och vattendrag tar emot stora mängder organiskt material från omgivande avrinningsområden. Det organiska materialet bildas vid nedbrytning av växter, till exempel i skogsjordar och myrmarker. Bakterier i sjöar och vattendrag utnyttjar detta organiska material som en energikälla, på samma sätt som vår egen användning av organiskt material i födan. Bakterier återinför därmed det döda organiska kolet till näringskedjan. Insikten om betydelsen av dessa processer på 1980-talet medförde ett paradigmskifte inom limnologin. Istället för att som tidigare fokusera på organiska näringsämnen, kväve (N) och fosfor (P), som basen för de biologiska processerna i sjöar och vattendrag, ändrades fokus till att också studera organiskt material, särskilt kol (C), från avrinningsområdet som en hörnsten i ekosystemet.

Det står nu klart att det organiska kolet från land mineraliseras i stor utsträckning i ytvattnen, huvudsakligen via bakteriernas respiration. Detta medför att koldioxid frisätts och avgår till atmosfären. Under syrgasfria förhållanden frisätts även metan. Bakteriernas aktivitet gör att vattnet övermättas med dessa båda växthusgaser, som därmed avgår till atmosfären. Även solstrålning bidrar till mineraliseringen av organiskt kol i ytvattnen genom fotokemisk mineralisering. Kunskap om dessa processer är av stort intresse både för forskning och för samhället i stort. För det första är det viktigt att känna till de naturliga processernas omfattning i relation till den pågående ökningen av koldioxid i atmosfären. Det är även viktigt att utröna hur processerna i ekosystemen kan förväntas förändras i samband med pågående och kommande miljöförändringar. Vattnets väg genom landskapet ändras i snabb takt genom omfattad byggnation av dammar, genom skogsbruk och ytterligare vattenverksamheter, vilket har effekter på frisättningen av växthusgaser från ytvattnen till atmosfären. Dessutom försvårar det organiska kolet möjligheterna att använda sjövattnet som resurs till dricksvattenförsörjningen, bland annat genom effekter på bakterietillväxt i ledningsnätet, produktion av giftiga ämnen vid behandling med klor för att kontrollera mikroorganismer, och genom att ge vattnet en brunaktig färg. Detta och andra effekter av det organiska materialet på sjövattnets kvalitet har direkt betydelse för samhället.

I denna avhandling utnyttjar jag det organiska materialets optiska egenskaper (absorbans och fluorescens), dvs dess färgegenskaper, för att undersöka dess koncentration och sammansättning. Båda dessa egenskaper har betydelse för hur det organiska materialet omsätts, och hur det bidrar till produktionen av växthusgaser. Dessutom är möjligheterna att avlägsna det organiska materialet vid rening av dricksvatten beroende av sammansättningen, som därmed påverkar vilka behandlingsmetoder som är lämpliga. Med denna bakgrund har min avhanding tre utgångspunkter. För det första, en bättre förståelse av variabiliteten i ytvattnens färg leder till en bättre kunskap om det organiska materialet. Nya mätinstrument utvecklas för närvarande snabbt, vilket gör det möjligt att mäta färgändringar i ytvattnet direkt i fält med hjälp av automatiska mätstationer, på ett sätt som aldrig gjorts tidigare. För det andra, olika signaler i vattens färg relaterar till olika aspekter av det organiska materialets kvalitet. För det tredje, transporten av organiskt material från land till sjö medför ett nära samband mellan det organiska materialet i jordar, i avrinningsområdet och i sjön. Sjöarnas omsättningstid, dvs hur länge vattnet uppehåller sig innan det transporteras vidare nedströms, spelar en stor roll för organiska materialets kvantitet och kvalitet.

Flera tusen sjöar från sju stora svenska avrinningsområden (Lagan, Mälaren, Dalälven, Ljungan, Ångermanälven, Umeälven och Kalixälven) analyserades för att bestämma sjöarnas omsättningstid. Majoriteten av dem byter helt ut sitt vatten under inom ett år. Sjöarnas omsättningstid bestäms till stor del av vårflöden i samband med snösmältningen och av nederbörd under sommar och höst. Med ökande omsättningstid i sjön minskar det organiska materialets färg, varvid fotokemisk nedbrytning till följd av solljus spelar en stor roll. Denna färgminskning är väl känd och sker främst under de första månaderna efter att löst organiskt material importerats från land till sjö. Mindre känd är signalen som relaterar till nybildning av organiskt material som sker internt i sjöar.

Vi genomförde en intensiv mätkampanj av absorbans vid en produktiv slättlandssjö, Tämnaren i Uppland (60°10’N, 17°20’E), för att undersöka förändringar i det organiska materialet under den produktiva sommartids-perioden, och med hög tidsupplösning (mätning var 30:e minut under flera månader, inom en stor del av det ultravioletta-synliga ljuset, 254-420 nm). Resultaten visar att den interna nyproduktionen av organiskt material inte registreras med absorbansmätningen, medan fluorescensmätningen hade förmågan att göra det. Fluorescensmetodernas förmåga att registrera den interna nybildning av organiskt material i sjöar är därmed en fördel över de klassiska absorbansmätningar där signalförluster oftast uppstår även vid den interna nybildning av organiskt material. Anledningen till detta förhållande kan å ena sidan vara en låg absorbansförmåga av det nybildade organiska materialet ifrån vissa källor. Å andra sidan förstör solen dem absorberande organiska komponenterna effektivt under den produktiva sommartidsperioden.

Denna observation stöds med en detaljerad analys av fluorescensmätningar ifrån flera hundratals sjöar. Analysen avslöjar sex olika fluorescens-komponenter som ge olika information om kvalitet av det lösta organiska materialet, och som relateras till olika egenskaper av vattnets kvalitet (t.ex. järn-innehåll, pH, och färg).

Sammanfattningsvis, visar min avhandling att absorbans ger en god bild av mängden organiskt material som importeras från land till insjöar. Dessutom är det relativt enkelt att mäta, och att automatisera. För den automatiserade mätningen av absorbans i sjöar ges detaljerade tekniska råd till genomförandet som anses tillämpbara även i grumligt sjövatten.

Jag föreslår användning och vidareutveckling av väl kalibrerade fluorescensmetoder för syftet att registrera nybildning av organiskt material som sker i sjön. Att registrera denna nybildning är viktigt då sjöns metabolism misstänks driva en snabb omsättning av lätttillgängliga och näringsrika organiska komponenter, t.ex. proteiner, vilket leder till relevanta kolflöden. Dessutom förändrar dessa nybildning också kvaliteten av det lösta organiska materialet vilket påverkar vattnets kvalitet, särskild i samband med dess behandlingsbarhet under dricksvatten förberedelse.

Thank you

This PhD project has been a wonderful opportunity for me, and I have enjoyed it endlessly. And so, for that, I would like to thank the many people who played a part in making my experience here so memorable.

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