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LETTER

Synchronous variation of dissolved organic carbon in Adirondack lakes at multiple timescales

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Scientific Significance Statement

Dissolved organic carbon (DOC) has significant effects on lake ecosystem function and in some regions exhibits upward trends referred to as "brownification." However, long-term trends typically account for minor variability in DOC and patterns and mechanisms of seasonal, annual, and multiannual variation are less understood. Synchronous fluctuations in DOC among lakes indicate common drivers that potentially affect variation. We document synchronous fluctuations in DOC across lakes recovering from acidification in Adirondack Park, New York, USA, that vary in strength across timescales. Relationships with external drivers and lake chemistry also differed by timescale: for example, higher precipitation was associated with lower DOC concentrations on seasonal timescales, but increased DOC on annual timescales. These findings indicate the importance of varying timescale-dependent mechanisms in lake DOC fluctuations.

Abstract

Dissolved organic carbon (DOC) is a key component of aquatic ecosystems with complex effects on ecosystem function. While long-term increases in DOC termed "brownification" have received considerable attention, directional trends typically account for a minority of variance. DOC concentrations also fluctuate on seasonal to multiannual timescales, but the causes of such variations are less understood. We used a wavelet-based approach to study timescale-specific, spatially synchronous fluctuations in DOC across 49 lakes in the Adirondacks, New York, USA. DOC varies synchronously among lakes at within-season, annual, and interannual timescales, but relationships with external drivers and internal processes indicated by lake chemistry differed across timescales. External drivers explained 78% of spatial DOC synchrony at the annual time scale. Beyond positive trends related to regional recovery from acidification, variability in DOC is a consequence of fluctuations at several timescales that are common among Adirondack lakes in precipitation, solar radiation, and internal chemical concentrations.

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Data Availability Statement: All data used in this study were obtained from publicly available sources cited in the manuscript. Versions of datasets analyzed in this study and analysis code have been archived on Zenodo. The URL is https://doi.org/10.5281/zenodo.7653396.

Additional Supporting Information may be found in the online version of this article.

Lakes in the northeastern United States have undergone recovery from acidification as environmental policy has reduced emissions that cause acid deposition (Driscoll et al. 2016). The rebound in lake and soil pH has resulted in increases in acid neutralizing capacity, less acidic pH, and higher concentrations of dissolved organic carbon (DOC; Driscoll et al. 2016; Bukaveckas 2021). Changes in these concentrations are of special interest because of the effects of DOC on vertical attenuation of light, water column temperature, and as a carbon source supporting food webs (Solomon et al. 2015). At low concentrations increasing DOC increases primary productivity because nutrients accompany DOC inputs, but at high DOC concentrations productivity declines due to reduced light availability (Solomon et al. 2015; Isles et al. 2021). Consequently, there has been widespread interest in regional browning trends and thresholds where effects of DOC on lake ecosystems change (Seekell et al. 2015; Weyhenmeyer et al. 2016; Williamson et al. 2016).

Although long-term trends in biogeochemistry associated with recovery from acidification have received considerable research attention, long-term trends account for only around one-tenth to one-fifth of the temporal variance in DOC (Bukaveckas 2021), leaving variations around these trends understudied (Strock et al. 2016; Imtiazy et al. 2020). Aquatic ecosystems vary on seasonal to multiannual scales due to external drivers interacting with internal processes (Wilkinson et al. 2020). The significance of oscillations in DOC include crossing thresholds from enhancement to inhibition of primary production. Timescale-specific (equivalently, frequency-specific; timescale is 1/frequency) approaches including wavelet analysis can identify and quantify these patterns and improve inference into pattern-generating mechanisms (Sheppard et al. 2016; Anderson et al. 2021).

Spatial synchrony provides a lens into the causes and consequences of fluctuations about trends over a range of timescales. Spatial synchrony (hereafter, "synchrony") describes when fluctuations in a focal variable are correlated across multiple locations. Long studied in population dynamics (Moran 1953; Liebhold et al. 2004; Walter et al. 2017), interest in synchrony in ecosystem processes has recently grown (Imtiazy et al. 2020; Kominoski et al. 2020; Walter et al. 2020; Seybold et al. 2021), in part because fluctuations that are synchronized across locations dominate total variability at larger spatial scales. This is because synchronous fluctuations reinforce each other in the aggregate, while asynchronous fluctuations tend to cancel each other (Anderson et al. 2021). Consequently, the mechanisms of synchrony are often major drivers of large-scale temporal variation. However, studies of lakes have found little synchrony at macro scales (Soranno et al. 2019) and highly variable synchrony (Lottig et al. 2017), even in ensembles of lakes separated by a few km (Baines et al. 2000), possibly in part because the measures used in these studies are confounded by timescale-specific dynamics. Recent studies demonstrate how patterns of spatial synchrony and their causes can differ across timescales and provide statistical methods that improve pattern detection and inference of mechanisms (Sheppard et al. 2016, 2017; Haynes et al. 2019; Walter et al. 2019).

In this study we analyzed long-term water chemistry data from 49 lakes in the Adirondack region of New York, USA, focusing on the following questions: (1) What are the spatial and timescale-specific structures of synchrony in lake DOC concentrations? (2) What is the relationship between synchronous DOC fluctuations and synchrony in (a) other biogeochemical constituents and (b) hypothesized external environmental drivers of DOC variation? (3) What fraction of synchrony in DOC concentrations is explained by coherent external drivers at different timescales? Our study reveals how DOC fluctuations in lakes recovering from acidification differ across timescales and depend on both external drivers and internal processing.

Methods

Study area

We used data from the Adirondack Long-Term Monitoring (ALTM) program that sampled lakes from 1992 to 2017 (Driscoll et al. 2003). These lakes lie in the Adirondack Park of New York State (Fig. 1), which covers > 2.4 million ha of public and private land. The lakes are mainly oligotrophic in largely undeveloped watersheds surrounded by wetlands and forests. Lakes were selected for long-term monitoring based on a > 500 lake survey in the 1980s (Kretser et al. 1989) and a subsequent classification of these lakes into seven categories based on hydrological and chemical properties (Newton and Driscoll 1990). Lakes in the ALTM study varied in area from 0.8 to 512 ha, in maximum depth from 1.2 to 24.4 m, and in mean DOC from 2.0 to 15.8 mg L^{-1}. Although there is evidence of recovery from acidification across the region, the degree of acidification and recovery rates depend on underlying geology (Driscoll et al. 2016). In the study area, most DOC is sourced from upland forests and estimated net in-lake DOC production is generally low, that is, < 1 kg C ha^{-1} yr^{-1} (Canham et al. 2004).

Data

Our primary dataset was the ALTM program (Driscoll et al. 2003). We extracted measurements of DOC, dissolved inorganic carbon (DIC), fluoride (F^-), chloride (Cl^-), sulfate (SO_4^{2-}), nitrate (NO_3^-), potassium (K^+), calcium (Ca^+), ammonium, (NH_4^+), magnesium (Mg^+), sodium (Na^+), acid neutralizing capacity (ANC), total dissolved aluminum, and inorganic monomeric aluminum. Analytical methods are reported in Driscoll and Van Dreason (1993). We excluded three lakes that were limed to increase their pH. We analyzed 49 lakes sampled monthly, year-round from June 1992 to September 2017 (> 26 years). Less than 1% of observations were missing and these were not concurrent across many

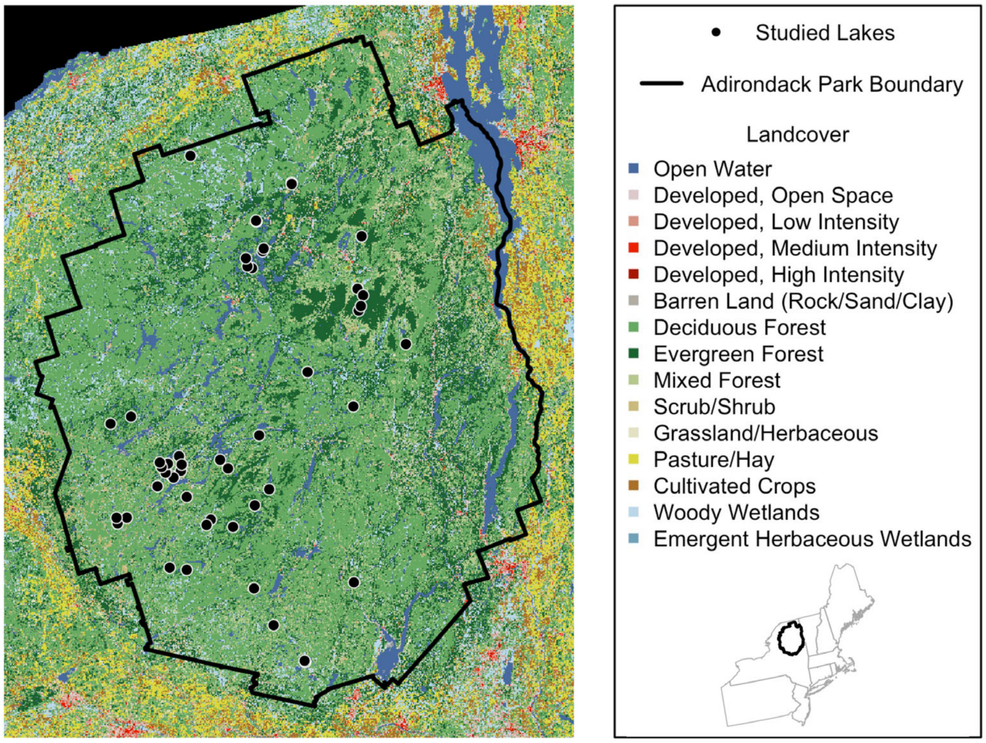

Fig. 1. Study area map. The locations of studied lakes and the Adirondack Park boundary overlay the National Land Cover Dataset (NLCD) landcover classification (year 2011 conditions).

lakes. Missing values were filled with the median across years for that lake and month. Thirty-three anomalous DOC values likely arising from laboratory error (i.e., DOC $<0.5\ \text{mg L}^{-1}$ and DOC $>25\ \text{mg L}^{-1}$) were also replaced with lake- and month-specific medians.

We obtained data on precipitation, the North Atlantic Oscillation (NAO) climate mode, deposition of sulfate and nitrogen, and incident photosynthetically active and ultraviolet (UV) radiation. Monthly total precipitation was extracted from PRISM (PRISM Climate Group 2020). Monthly NAO indices were obtained from the NOAA Climate Prediction Center (https://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/nao.shtml). Monthly atmospheric deposition data were obtained from the National Acid Deposition Program (NADP) National Trends Network (https://nadp.slh.wisc.edu). We averaged time series for eight monitoring stations located within or very near the Adirondack Park into a representative acid deposition time series for the study area. Data for spectrally resolved solar surface irradiance at each lake were taken from The National Solar Radiation Database's Spectral On-Demand service for the sampling sites, which uses the Fast All-sky Radiation Model for Solar applications with Narrowband Irradiances on Tilted surfaces (Xie et al. 2018; Xie and Sengupta 2018). The irradiance data cover 280–4000 nm at hourly time steps beginning in January 1998. The UV band spanned 280–300 nm and the photosynthetically active radiation (PAR) band spanned 400–700 nm. Both were aggregated to monthly means to match the lake data.

Prior to analysis, each time series was detrended, transformed to approximately normal marginal distributions using a Box–Cox transformation, and standardized to have mean $= 0$ and standard deviation $= 1$. This procedure prepares the data to match assumptions of our statistical tests.

Analyses

We performed two explorations of geographical structures in the synchrony of DOC (Supporting Information S1); finding little, we henceforth focused on analyses of the timescale structure of synchrony in DOC and how DOC synchrony is related to other aspects of lake chemistry and to external drivers expected to shape inputs (e.g., precipitation) or losses (e.g., acid deposition, irradiance) of DOC.

To characterize how synchrony in DOC among Adirondack lakes changed through time and depended on timescale of variation, we used wavelet phasor mean field analysis (Sheppard et al. 2016). Wavelet phasor mean fields quantify phase synchrony, in this case, the tendency for fluctuations in DOC to be temporally aligned across lakes, allowing for variability across timescales and through time. Synchrony was significance tested against a null model of no synchrony in which synchrony arose only by chance, as represented by 10,000 surrogate sets of 49 random phasors. We calculated p-values based on the quantile of empirical phase synchrony relative to that of surrogates; we considered synchrony statistically significant at the 0.001 level if the empirical synchrony exceeded 99.9% of surrogates.

To test for timescale-specific, synchronous relationships between DOC and lake chemistry and external drivers, we used spatial wavelet coherence (Sheppard et al. 2016). Spatial wavelet coherence tests whether two spatially resolved variables fluctuate with related magnitudes and consistent phase differences, across locations, as a function of timescale. Spatial wavelet coherence produces a magnitude quantifying the strength of association and a phase indicating whether the relationship is positive, negative, or time lagged. Statistically significant wavelet coherence between a response variable and an external driver can be interpreted as evidence that the external driver is a causal mechanism of synchrony in the response (Sheppard et al. 2016; Walter et al. 2019; Anderson et al. 2021). Variables such as precipitation and UV radiation can be regarded as external drivers; for most lake chemistry variables, such as cation or anion concentrations, the direction of causality, or whether coherence with DOC is caused by some shared third driver, is uncertain. Significance testing was performed using the "fast" method of Sheppard et al. (2017), which tests the observed coherence magnitude against that of surrogates representing a null hypothesis of no coherence that preserve the spatial and temporal autocorrelation properties of the original variables, but in which coherence between the two variables arises only by chance. We measured spatial wavelet coherences between DOC and all considered variables over three timescale bands: seasonal (2–4 months), annual (9–16 months), and interannual (2–8 yr). To visualize relationships between DOC and water chemistry and environmental drivers, we used box-and-arrow diagrams which connected DOC with variables with statistically meaningful coherence relationships. Note that due to the solar radiation data beginning in 1998, for analyses with solar radiation the longer time series were truncated to the shorter period.

To quantify the fraction of synchrony in DOC attributable to relationships with external drivers, we applied wavelet linear models and the synchrony attribution theorem (Sheppard et al. 2019). Wavelet linear models extend pairwise coherence testing as performed above to models that account for the effects of multiple predictors, analogous to the difference between correlation and multiple regression. We built wavelet linear models using as predictors all external driver variables that were substantially coherent ($p < 0.1$) with DOC at a timescale band, except if two predictors strongly covary in which case one of the strongly covarying predictors was omitted. ANC was considered an external driver, alongside NAO, precipitation, acid deposition variables, and solar radiation variables. Although lake primary production could influence ANC, available observations indicate the effect is weak in these lakes (Pearson correlation = −0.08). Solar radiation variables were highly coherent (Supporting Information S2), so we chose the more coherent one and present alternative results in Supporting Information.

Analyses were conducted in R version 4.1.1 (R Core Team 2020) using the "ncf" and "wsyn" (Reuman et al. 2021) packages.

Results

Lake DOC varied through time and among Adirondack lakes (Fig. 2a), with interannual variability generally a moderate fraction of overall variability (Supporting Information S3). Major temporal variations emerge in the average (Fig. 2b), which is heavily influenced by synchronous fluctuations. The wavelet phasor mean field diagram (Fig. 2c) illustrates the degree of synchrony and its variability across time and

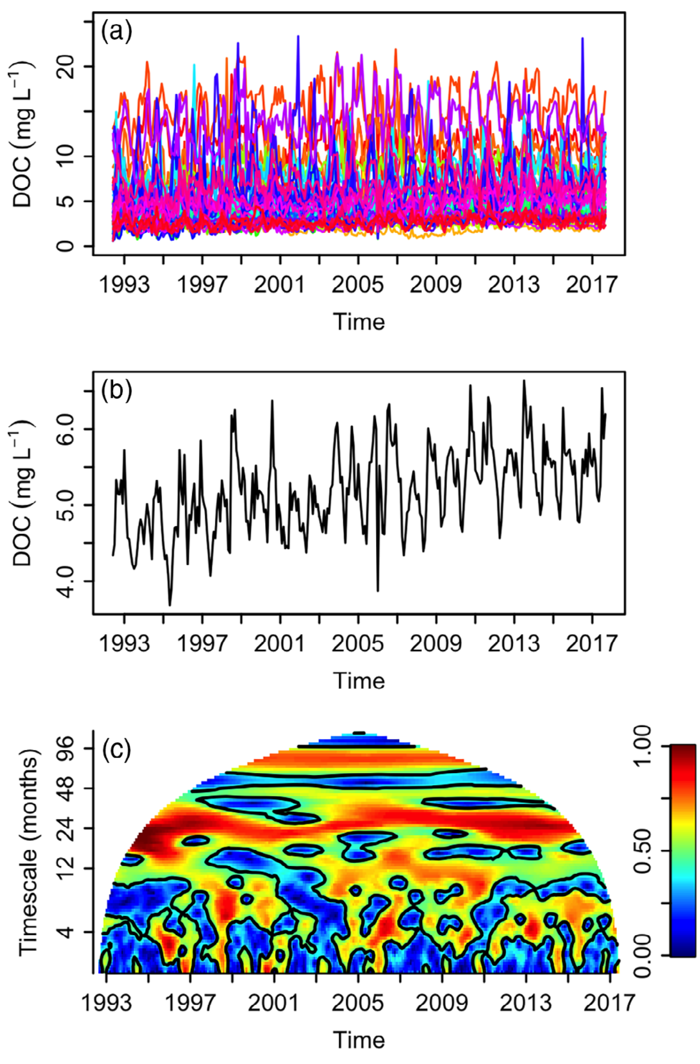

Fig. 2. (**a**) DOC time series from 49 Adirondack lakes. (**b**) Mean DOC time series across 49 lakes, which primarily reflects synchronous fluctuations shared across the lakes. (**c**) Spatial synchrony of DOC fluctuations depends on time and timescale, as quantified using the wavelet phasor mean field. Black contours indicate statistically significant synchrony ($p < 0.001$), which correspond to the areas on the colored gradient from green to red.

timescale. Within-season synchrony includes distinct episodes (red regions) in 1996–1997 and 2005–2006 that were likely driven by meteorological events (Fig. 2c). Annual synchrony was prevalent, but conspicuously low (blue band) ca. 1999–2002. At interannual timescales, there was a high degree of synchrony in DOC at about 2 yr, and at approximately 7 yr.

Spatially coherent relationships differed among the seasonal (2–4 months), annual (9–16 months), and interannual (2–8 yr) timescales, as indicated by the changing presence/absence of links between variables, and their changing colors, which indicate their phase relationships (Fig. 3).

On seasonal (2–4 months) timescales, DOC was negatively associated with precipitation, suggesting that the dominant effect at very short timescales is dilution (Fig. 3a). ANC showed an antiphase (negative) relationship with DOC, possibly because DOC is weakly acidic, while ANC is mainly driven by inputs of strong acids. The combination of precipitation and ANC explained 12.3% of synchrony in DOC; precipitation was the primary synchronizing driver (9.8% of synchrony explained by precipitation). There were a mix of in-phase (positive) and antiphase (negative) relationships between DOC in major cations and anions, and strong in-phase (positive) relationships with aluminum compounds linked to fish toxicity.

On annual (9–16 months) timescales, precipitation was a temporally lagged driver of lake DOC concentrations, consistent with DOC inputs being driven by precipitation but lagged by hydrologic transport times. At annual timescales, other external drivers were also important (Fig. 3b). Note that some correlation is expected at these timescales due to seasonal cycles; however, our significance testing procedures preserve the spectral structure of the original time series, thus significance implies that variability in the magnitude or phase offset of these cycles is correlated across years. DOC and acid deposition had antiphase (negative) relationships. Solar radiation variables (UV and PAR) were considered due to their role in photodegradation of DOC, but their phase relationship, which indicates that DOC increases while solar radiation is above average, is inconsistent with this expectation. These external drivers explained 79.5% of synchrony in DOC (Fig. 4). Note that we observed net negative interaction effects between driver variables; in other words, observed DOC synchrony was less than expected if all driver variables acted independently (Sheppard et al. 2019; Castorani et al. 2022). Results differed modestly when PAR replaced UV radiation in the model (Supporting Information Fig. S4). Like seasonal timescales, relationships between DOC and major anions and

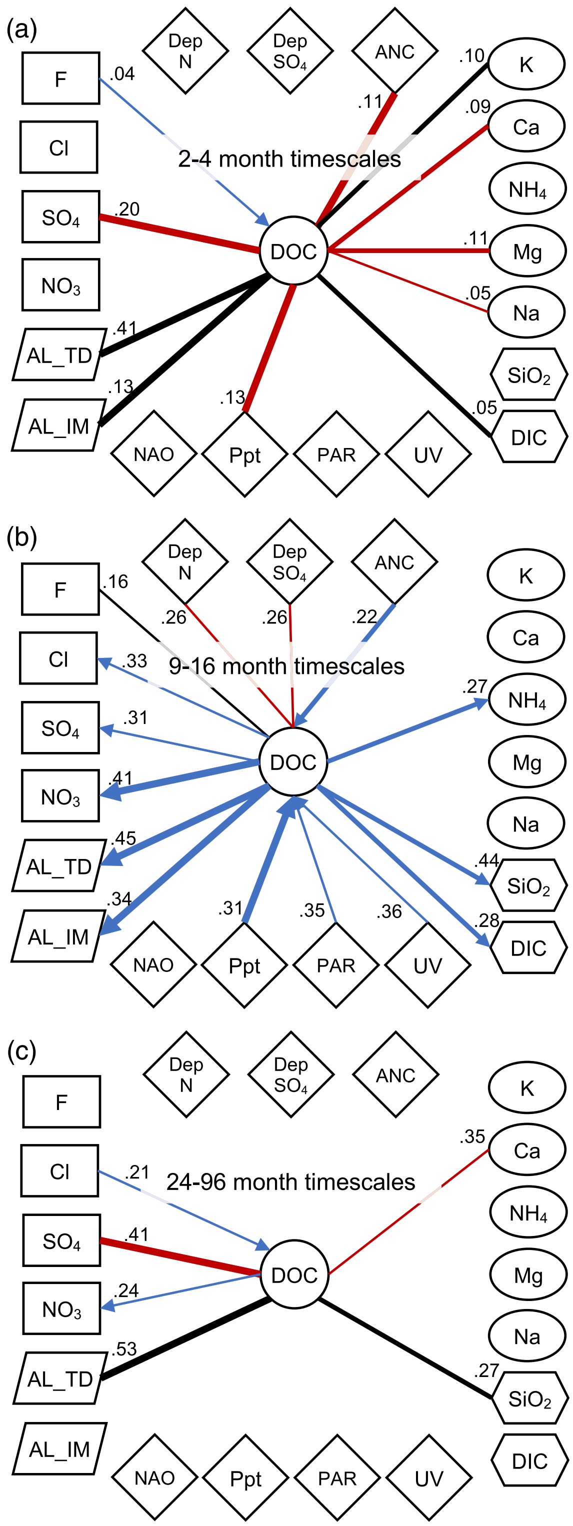

Fig. 3. Spatial wavelet coherence relationships at (**a**) seasonal (2–4 months), (**b**) annual (9–16 months) and (**c**) interannual (2–8 years) timescales. Line width indicates significance level: 1 pt. = $p < 0.1$; 2 pt. = $p < 0.01$; 3 pt. = $p < 0.001$. Line color indicates class of phase relationship: black = approximately in-phase; red = approximately antiphase; blue = time lagged, where the arrowhead points toward the lagging variable. Numbers indicate coherence magnitudes; note that due to a bias toward larger values at longer timescales these should be interpreted as comparisons of coherence strength within a timescale band only. Box shapes group similar variables, for example, diamonds are external drivers, rectangles are anions, and ovals are cations. "Ppt" = total precipitation; "Dep N" = nitrogen deposition; and "Dep SO_4" = sulfate deposition.

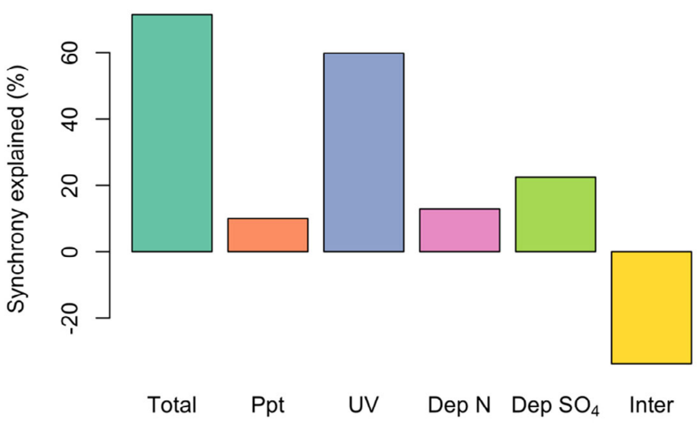

Fig. 4. Synchrony explained by external drivers at 9–16 months timescale. "Ppt" corresponds to total precipitation; "Dep N" corresponds to nitrogen deposition; and "Dep SO_4" corresponds to sulfate deposition. "Inter" indicates the sum of two-way interactions between predictor variables; the negative interaction term indicates that synchrony in DOC is reduced by interacting effects of driver variables.

cations varied in strength and phase, and there were strong in-phase relationships with aluminum compounds. There was also a strong relationship in which SiO_2 lagged DOC.

At interannual (2–8 years) timescales, relationships between DOC and environmental drivers were not apparent (Fig. 3c). Since no factor exhibiting a significantly coherent relationship to DOC could clearly be considered an external driver, we did not measure the synchrony in DOC explained at this timescale band. However, DOC remained linked to some major cations and anions, to aluminum compounds, and to SiO_2.

Discussion

Concentrations of DOC in Adirondack lakes exhibited timescale-specific, spatially synchronous oscillations that can be attributed to external drivers and internal processes. Although DOC synchrony showed little discernible geographic structure, similar to other studies of synchrony in lake ecosystem dynamics (Lottig et al. 2017; Soranno et al. 2019), our finding of strong synchrony in DOC variation at seasonal, annual, and interannual timescales is notable. Synchronous variation among lakes across the Adirondacks indicates common processes, especially shared external drivers, largely determine DOC dynamics. This was especially true for annual-scale oscillations, where external drivers explained >70% of synchrony in DOC. The strength and phase of relationships between DOC, external drivers, and lake chemistry differed across timescales; non-timescale-specific approaches could not resolve this, which would lead to undetected or mischaracterized effects. In addition to long-term trends, variation in DOC is related to dynamics at seasonal, annual, and multiannual time scales. Because of how DOC influences light, temperature and nutrients in lakes, and how current DOC concentrations influence whether subsequent changes promote or inhibit primary production, timescale-specific oscillations in DOC have important implications for lake ecosystem dynamics.

Synchronous variation in DOC concentrations was driven partly by variation in precipitation on seasonal to annual time scales. Considering that the effects of precipitation variation on lake ecosystems may differ by hydrologic setting (Oleksy et al. 2022) and Adirondack lakes are a mix of drainage and seepage lakes, our finding that precipitation drove synchronous DOC variation is perhaps surprising. Studying lakes in the same region, Strock et al. (2016) found that DOC concentrations increased in years of above normal precipitation and declined in years of below normal precipitation. The lakes in this study are oligotrophic, so upland forests are the dominant source of DOC (Canham et al. 2004), which is delivered to lakes via flushing from soils during snowmelt and precipitation events (Boyer et al. 1997; Solomon et al. 2015; Strock et al. 2016). Our finding that phase relationships differed between timescales, suggesting dominant mechanisms shifted from dilution due to large precipitation events at within-season (2–4 months) timescales to lagged delivery possibly originating from seasonal snowmelt at annual (9–16 months) timescales. The time lags between peaks in precipitation and DOC at annual timescales are generally consistent with rates of hydrologic transport through soils (Lambert et al. 2014).

At annual timescales, DOC dynamics were related to the external drivers of acid deposition (NO_3 and SO_4) and solar radiation (UV and PAR). Although nitrogen deposition has been linked to greater export of humic material from watersheds (Porcal et al. 2009), we observed an antiphase (negative) relationship more consistent with increases in acidity, particularly as SO_4, increasing photochemical oxidation rates of DOC in lakes and reduced soil solution DOC concentrations (Porcal et al. 2009). In the study lakes, NO_3 and SO_4 concentrations were positively correlated, suggesting that shared mechanisms could drive temporal fluctuations in each. Although we expected that increases in solar radiation would increase rates of photodegradation of DOC in lake waters, we observed lake DOC to increase while solar radiation was above average. One possible explanation is that increasing solar radiation tends to increase the amount of carbon fixed by forests and rates of biogeochemical processing and hydrologic transport account for DOC lagging solar radiation. Forest production in this region may be limited more by sunlight and temperature than water (Boisvenue and Running 2006). Although it is possible that DOC and any variable exhibit coherent annual-timescale cycles due to some exogenous mechanism, shared seasonality is not sufficient to produce significant coherence because our significance testing procedures account for the cycles inherent in both variables.

Concentrations of DOC also exhibited several relationships with other aspects of lake chemistry, including major cations and anions, aluminum, DIC, and silica (SiO_2). Some were expected given known mechanistic relationships; for example, DOC enhances the bioavailability of aluminum (DeForest et al. 2018), and DOC and two aluminum species were consistently positively related across timescales. Although not all mechanistic relationships are clear, lagged relationships are suggestive of coupling through internal biogeochemical processes. If the shared action of external drivers alone drove coherence between DOC and other aspects of lake chemistry, we should expect in-phase (positive) and antiphase (negative) relationships to predominate. Instead, lagged relationships, which suggest processes that degrade and transform lake inputs, were about equally common. However, lags could also result from differences in mobility through the watershed. Further supporting a combined role for external drivers and biogeochemical processes, at interannual timescales there were no coherent relationships between DOC and external driver variables, but a handful of strong relationships between DOC and components of lake chemistry remained.

Our study revealed how the synchronized lake water chemistry fluctuations of a region recovering from acidification differ across within-season to interannual timescales, and how this is related to both external drivers and internal biogeochemical processes. Timescale-specific dynamics, especially fluctuations having multiyear period lengths, are understudied in aquatic ecosystems (Wilkinson et al. 2020), can lead mechanistic relationships to be undetected or mischaracterized using conventional non-timescale-specific tools, and likely manifest in many other contemporary dynamics of inland waters related to long-term processes like eutrophication and salinization. The approaches used here provide a roadmap for characterizing spatiotemporal patterns of these phenomena—especially fluctuations shared across many water bodies or sampling locations—and beginning to resolve their mechanisms. This and other recent studies (Imtiazy et al. 2020; Kominoski et al. 2020; Walter et al. 2020; Seybold et al. 2021) demonstrate that synchrony can be a powerful lens for ecosystem science and biogeochemistry.

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