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Macrophyte cover type and groundwater as the key drivers of the extremely high organic carbon concentration of soda pans

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Summary

1. Endorheic soda pans are among the aquatic systems that have the highest dissolved organic carbon (DOC) content on the planet with concentrations reaching values close to 1 g L^{-1}. Considering the importance of inland waters in the global carbon cycle, the understanding of the drivers of such outstanding aquatic organic carbon pools is eminent. The soda pans of the Carpathian Basin present a wide variability of biotic and abiotic characteristics that provides an adequate system to assess the determinants of the extreme high DOC concentrations of soda pans. Here we demonstrate through a multisite comparison, a multiyear seasonal monitoring, and a laboratory experiment that the dissolved organic matter content of the highest DOC concentration soda pans is primarily of groundwater and emergent macrophyte origin.
2. More precisely, the multisite comparison of 14 soda pans revealed that the variation of colored dissolved organic matter (CDOM) content of the pans is partially explained by the CDOM content (21% of variation) and conductivity (14%) of the local groundwater indicating significant role of allochthonous terrestrial DOC sources. However, 46% of the variation in CDOM content of the studied soda pans could be accounted for the dominant type of emergent macrophyte with *Bolboshoenus maritimus* dominated macrophyte cover leading to higher CDOM content than *Phragmites australis*.
3. In line with the results of the multisite comparison, we demonstrated by a decomposition experiment that both *B. maritimus* and *P. australis* have the potential to release substantial amount of organic matter into soda pans. However, the organic matter release of *B. maritimus* is much more intensive than that of *P. australis* leading to twice as high DOC and 3.5-times higher CDOM concentrations. In general, considering previous organic matter release studies we concluded that *P. australis* is a relatively low

organic matter releaser emergent macrophyte, and therefore the species composition of emergent macrophytes has to be considered in autochthonous plant-derived DOM estimations.

4. Finally, the multi-year seasonal monitoring of two distinctive soda pans showed that the high organic matter concentrations of the pans depends not only on their intrinsic characteristics but also on interannual variability. More precisely, we demonstrated that the highest CDOM and DOC concentrations occurred in a colored (i.e., brown, low TSS) soda pan that had extensive (95%) macrophyte cover dominated by *B. maritimus* in a period characterized by high pH due to low water levels, which were presumably the consequence of increased evaporation due to decreased precipitation and above average temperature. Considering the trends of climate change in Central-Europe (i.e., increased temperature and modified precipitation regimes), our results indicate that extremely high organic matter concentrations in soda pans might become more frequent in the near future.

Keywords: high pH, DOM, groundwater effect, emergent macrophytes, interannual variability

Introduction

As inland waters receive, process, store and emit carbon in globally significant amounts (Cole *et al.*, 2007), mapping active carbon in even the smallest aquatic systems is necessary for accurate global estimates of carbon cycling (Tranvik, Cole & Prairie, 2018). Dissolved organic matter (DOM) is quantitatively the most significant pool of organic carbon in aquatic systems, which is either derived from terrestrial sources (i.e., allochthonous organic matter) or from biological material produced *in situ* by phytoplankton and macrophytes (i.e., autochthonous OM) (Williamson *et al.*, 1999; Zhang *et al.*, 2013). Terrestrial DOM can reach surface waters via surface inflow or runoff as well as via groundwater seepage (Wetzel, 2001; Grabs *et al.*, 2012; Einarsdottir, Wallin & Sobek, 2017). Therefore, the quantity and quality of DOM in inland waters is not only influenced by internal processes but also by the characteristics of the catchment area (e.g., vegetation, soil type, hydrology) (Wetzel, 2001; Kothawala *et al.*, 2014; Sepp *et al.*, 2019).

In inland waters DOM is considered to be dominated (15–80%) by soluble humic substances (i.e., fluvic acids and humic acids). While fulvic acids are soluble at any pH, the solubility of humic acids depends on pH with no dissolution at lower pH (pH < 2) and complete solubility at pH 13 (Aiken *et al.*, 1985). The pH of the water also affects the properties of the dissolved humic substances with more aromatic and aliphatic humic acids being dissolved at higher pH (Baglieri *et al.*, 2014). Humic acids also contribute to the fluorescence signal of colored (chromophoric) DOM (CDOM) (Lapierre & Frenette, 2009). Generally, allochthonous DOM is composed of more recalcitrant and highly colored humic substances than phytoplankton produced autochthonous DOM (Tranvik L. J., 1988; Wetzel, 2001). However, the refractory plant-derived CDOM can also originate from autochthonous sources such as littoral marshland vegetation (i.e., emergent macrophytes) (Lapierre & Frenette, 2009). Humic substances are considered to be recalcitrant due to their aromatic core, which is relatively resistant to microbial degradation (Kellerman *et al.*, 2015). However, it has been shown that terrestrial carbon can be substantial contributor to microbial biomass (Guillemette, McCallister & del Giorgio, 2015).

The global mean of dissolved organic carbon (DOC) concentration of lakes has been determined to be 5.02 mg L^{-1} (equivalent of 5.58 mg L^{-1} total organic carbon) and influenced by climatic factors (Chen et al., 2015) as well as watershed characteristics (Sobek et al., 2007). Although the globally most abundant shallow lakes (Downing *et al*., 2006; Verpoorter *et al.*, 2014) have been shown to have double mean DOC concentration than deep lakes (6.56 and 3.12 mg L^{-1}, respectively) (Chen et al., 2015), the 90 mg L^{-1} mean DOC concentration of soda lakes can be considered as extreme even when compared to other highly productive aquatic systems such as eutrophic lakes, marshes or bogs (Fig. 1a). In addition, when looking at single measurements, soda pans are very likely the global record holders in DOC as occasionally DOC concentrations close to 1 g L^{-1} have been reported (Lake Nakuru: 980 mg L^{-1} (Jirsa *et al.*, 2013); Sósér: 988 mg L^{-1} (Boros *et al.*, 2016) (Fig. 1b).

Soda lakes and pans can be found on all continents except Antarctica and represent the most alkaline natural environments on Earth (Grant & Sorokin, 2011). Soda lakes are formed in endorheic basins (i.e., limited drainage basins), where evaporation exceeds water outflow (Warren, 2006) and the levels of calcium (Ca^{2+}) and magnesium (Mg^{2+}) are low, while sodium (Na^{+}) and carbonate species (HCO_3^{-} + CO_3^{2-}) are high (Boros & Kolpakova, 2018). Within Europe—to the best of our knowledge—soda pans are restricted to the Carpathian Basin and are found only in Austria, Hungary and Serbia. The climatic conditions of the region (i.e., continental with influence of both oceanic and Mediterranean climate) in combination with the shallowness of the pans cause high fluctuation in respect of water level and temperature. Water level fluctuation is of particular interest as it results in intermittent hydroperiods for many pans and affects the concentration of both organic and inorganic compounds. In the case of soda pans of the Carpathian basin, groundwater inflow typically exceeds the surface-related watershed inflow and precipitation (Boros, Ecsedi & Oláh, 2013). The analysis of 84 soda pans of the region showed that many pans have extremely high DOC and CDOM concentrations (DOC: median = 47; range: 20–664 mg L^{-1}; CDOM: median = 310; range: 20–7,100 mg Pt L^{-1}), which indicates polyhumic (CDOM > 90 mg Pt L^{-1}) character (Boros *et al.*, 2017). Furthermore, extreme high DOC concentrations have also been recorded (Fig. 1b) (Boros *et al.*, 2016). Positive correlation between DOC and CDOM has been determined before for Carpathian soda pans (V.-Balogh et al., 2009; Boros et al., 2013; 2016; 2017), while—despite the pH-dependent solubility of humic substances—correlation between pH and DOC has not been demonstrated (Boros *et al.*, 2017).

Although high organic matter content is an inherent property of most soda lakes and pans, the regulators of CDOM variation across soda pans are understudied and the causes of extremely high DOC and CDOM concentrations such as those measured in the pans of the Carpathian basin, are not properly understood. Therefore, in this study we aim to identify and test the main sources of CDOM and DOC in polyhumic soda pans. Namely, we hypothesize that CDOM concentration of polyhumic soda pans is positively affected by the allochthonous CDOM concentrations of local groundwater and the extent of emergent macrophyte cover. Furthermore, based on our earlier observations (Boros *et al.*, 2017), we hypothesize that the species composition of the autochthonous emergent macrophytes influences CDOM content with cosmopolitan bulrush (*Bolboshoenus maritimus* (L.) Palla, Cyperaceae) dominated cover leading to higher CDOM concentrations than common reed (*Phragmites australis* (Cav.) Trin. ex Steud., Poaceae) dominated cover. To disentangle the effect of different macrophyte species on CDOM and DOC concentrations, experimental assessment of the amount and ratio of CDOM and DOC released from *P. australis* and *B. maritimus* was performed. Finally, to identify the environmental conditions that lead to extreme DOC and CDOM values in soda pans, seasonal monitoring of a turbid and a colored polyhumic soda pans was conducted in two separate years.

Methods

Study sites and sampling

The soda pans studied in this work are located in the central area of the Carpathian Basin, on the interfluve area of the Danube and Tisza rivers (Fig. 2). The water budget of the soda pans in this area is highly influenced by evaporation, precipitation and groundwater influx, while the surface water inflow from the watershed is negligible as usually no major watercourse enters these systems (Boros *et al.*, 2013). The primary source of the high Na–HCO_3–Cl^- content of the soda pans of the region is the discharge from upwelling deep saline groundwater, which is enhanced in the pans by evaporation (Simon *et al.*, 2011). These soda pans can be categorized into two groups based on their optical characteristics: the turbid type, where inorganic suspended solids (ISS) are the main cause of turbidity or more precisely, the contribution of ISS to light attenuation (K_d) exceeds 50%, and the colored (brown) type, where CDOM contribution dominates K_d (> 50%) (Boros *et al.*, 2013). Submerged and floating macrophytes are sparse or absent from the open water areas of both turbid and colored pans. However, marshland vegetation (*Bolboschoeno-Phragmitetum*) characterized primarily by varying ratios of emergent macrophyte species *B. maritimus* and *P. australis* can be found on their shoreline. This study comprises of three parts: a multisite comparison of soda pans, a decomposition experiment and a seasonal analysis of a turbid and a colored soda pan.

In the multisite comparison 14 natural pans were sampled between April and September of 2017 to assess the potential effect of groundwater and macrophytes on the organic matter content. The pans were selected in order to cover a broad range in respect of turbidity, CDOM and emergent macrophyte cover (Table 1). The coordinates of the location of the pans and the groundwater wells are listed in Table S1. The chemical type of the pans was determined following the guidelines of Boros & Kolpakova (2018). Two of the 14 pans were sampled in the seasonal analysis: Zab-szék, a typical turbid pan (Table 1: Total Suspended Solids (TSS) = 1574 mg L^{-1}, CDOM = 364 mg Pt L^{-1}) and Sósér, a typical colored pan (TSS 83 mg L⁻1, CDOM = 2088 mg Pt L^{-1}) (Boros et al., 2013; 2016; 2017). The seasonal assessment of these two pans was performed in 2014 and 2017 with samplings approximately every other week from January to December in 2014 and from March to November in 2017. On two occasions in 2017 (11-Sep and 16-Oct) Zab-szék was completely dry, so no measurement was possible.

For all samplings water depth, conductivity and pH were determined on site in the open water part of the pans using a centimeter-scale pole, and a WTW MultiLine P4 field instrument for pH and conductivity, respectively. Samples were collected and transported to the lab for CDOM, DOC, TSS, and fluorescence excitation emission matrix spectroscopy (EEMS) measurement. The groundwater was assessed by sampling dug wells (mean depth: 3–5 meters) located within 500 meters from the shoreline of each pan (Table 1). According to topographic maps, all of the sampled wells were established decades ago. For the macrophyte DOM release experiment, aboveground fraction of stems and leaves of *B. maritimus* and *P. australis* specimens in their early senescing stage were collected from Sósér soda pan in October 2012. After collection, the plant samples were washed with pan water and kept in clean plastic bags and brought to the laboratory.

Autochthonous emergent macrophyte cover assessment

The cover of open water and marshland vegetation (*Bolboschoeno-Phragmitetum*) of pans and the classification based on *P. australis* and *B. maritimus* ratio were estimated with remote-sensing databases extracted from Google Satellite raster data via OpenLayers plugin and complemented with local field observations in 2017. The GIS mapping procedure and spatial calculations were carried out using ArcMap (Environmental Systems Research Institute 2013). The proportion of emergent macrophyte cover (%) was calculated based on the ratio of open water and macrophyte cover (ha) and the characteristic macrophyte type of each soda pan was determined by estimation of the ratio of the most common species (*P. australis* and *B. maritimus*). Pans with ratio of either species above 50% were classified as either *P. australis* or *B. maritimus* dominated (Table 1).

Experimental release of DOM from macrophytes

In the laboratory the plant material was cut into 20-cm-long pieces, oven-dried to constant weight at 35°C to avoid the destruction of the associated microbiome, and stored at room temperature (23°C) until the experiment. Fifty grams of oven-dried plant material (stems and leaves) were placed into 5-liter bottles. The bottles were filled with 3.5 l water (pH = 8.4) collected from the well near Kelemen-szék (Table 1, N: 46.8012; E: 19.1717). The well-water was filtered with pre-combusted GF-5 acid-washed glass fiber filter (pore size = 0.4 μm). The DOC and CDOM concentration of the well water were determined as for other samples. The incubation was performed in three replicates in the dark at room temperature (22-24 °C) for 29 days. The bottles were aerated with sterile-filtered atmospheric air and the dissolved oxygen saturation ranged between 64% and 94% throughout the experiment. This aerobic treatment was chosen, because an earlier study on *P. australis* showed that aerobic conditions are more similar to the conditions in the field than anaerobic (V.-Balogh *et al.*, 2006). On days 0, 1, 4, 7, 11, 14, 18, 22, 25 and 29, 100 ml of water were sampled from the bottles for analyses. The water volume of the samples was always replaced, consequently the water volume remained constant during the experiment. All glassware used for sample collection and analytical processes was acid-washed and Milli-Q water rinsed.

CDOM, DOC, and TSS measurements

The water samples were filtered as for the DOM release experiment and the concentration of colored dissolved organic matter (CDOM) was expressed as Pt (platina) units (mg Pt L^{-1}) using absorbance (440 nm) measurements with a Shimadzu UV 160A spectrophotometer (Cuthbert & del Giorgio, 1992). For DOC analyses, the filtered samples were acidified (to pH 2 with HCl) and bubbled to remove dissolved inorganic carbon (DIC), and DOC concentration was measured by thermal catalysis at 1050 °C in an Elementar High TOC instrument equipped with a platinum cartridge using synthetic air as carrier gas. The concentration of total suspended solids (TSS) was measured by filtering water (100–2000 ml) through pre-dried and pre-weighed cellulose acetate filters (pore size = 0.45 µm) followed by oven-drying at 105°C, and weighing of dry filters using a CHYO YMC SM-200 analytical balance (accuracy 0.01 mg, precision 0.02 mg).

Fluorescence Excitation Emission Matrix Spectroscopy

During the 2017 seasonal comparison of Sósér and Zab-szék samples were collected for dissolved organic matter (DOM) characterization using fluorescence excitation-emission matrix spectroscopy (EEMS). The samples were filtered through a 0.1-µm pore size Millipore Isopore Membrane Filters the same day and stored in combusted glassware at 4°C until processing. EEMS profiles of the samples were determined as in Kothawala *et al.* (2014). Briefly, excitation-emission matrices (EEM) were determined by measuring UV-visible absorbance spectra using a Lambda 40 UV-visible spectrophotometer (Perkin Elmer, Waltham, MA, United States) and measuring fluorescence emission using a fluorescence spectrophotometer (SPEX FluoroMax-2, Horiba Jobin Yvon, Kyoto, Japan). Milli-Q water was used as blank and its values were subtracted from the EEMs. Manufacturer supplied instrument correction factors and the absorbance spectra were used for the correction of instrument and filter biases, respectively, while fluorescence intensity was calibrated to the Raman area of the blank water.

Data analyses

To assess the effect of macrophytes on the CDOM content of the pans, the amount of non-groundwater related CDOM (i.e., $CDOM_{diff}$) was calculated by subtraction of the CDOM concentration of the corresponding groundwater wells from the total CDOM concentration of pans.

From the EEMS results three indicators have been calculated: fluorescence index (FI), freshness index (FRESH), and humification index (HIX). The FI is used as an indicator of the source of DOM: high FI ~1.8 indicates microbial and algal origin of DOM, while low FI ~1.2 suggests terrestrial plant and soil derived DOM. The FRESH index is used as an estimator of how recently the DOM has been produced ('freshness'). Finally, HIX indicates the humification of DOM (i.e., amount of aromatic compounds) (Fellman, Hood & Spencer, 2010).

In the multisite comparison, the variables influencing CDOM concentration in the pans were evaluated by a linear model. The relevant variables for the linear model were selected from those in Table 1 (except the distance of the well to the corresponding pan) by backward selection using Akaike information criterion (AIC) estimator (Table S2). The linear model was checked for multicollinearity and the variables that had the highest generalized variance inflation factor (GVIF) were stepwise removed until reaching a model were the squared degrees of freedom corrected GVIFs ((GVIF^(1/(2*Df))^2) were less than five (Table S3). Differences of measured variables and linear models were tested by Welch's *t*-tests and Analyses of Variance (ANOVA), respectively. Correlations between parameters were evaluated with Pearson's correlation analyses. To meet the assumptions of normality of residuals of parametric tests and the requirements of linear correlations data was log-transformed when needed.

Normality of residuals and variables was checked by Shapiro-Wilk test and visual analysis. In the seasonal study the relationships over time of CDOM and DOC with TSS, water depth, pH, conductivity and the EEMS indexes were assessed on monthly averaged data using cross-correlation analyses (Zab-szék in 2017 was excluded from these analyses due to repeated complete droughts). Prior analyses the autocorrelation of the variables was removed by differencing the series with a lag of one. The successful removal of autocorrelation was confirmed by the Ljung-Box test. The Spearman correlation, non-linear curve fitting for Michaelis-Menten kinetic function, Mann-Whitney, and Kruskal-Wallis tests used to analyze the DOM release experiment were performed using OriginPro 9 (OriginLab, Northampton, MA), while the EEMs data were analyzed with MATLAB. All other analyses were performed using the R environment for statistical computing (R Core Team, 2015).

Results

Multisite comparison of the effect of groundwater and macrophytes

The identity, optical category (i.e., turbid or colored), chemical type (i.e., category based on ionic composition, Boros & Kolpakova, 2018), TSS as a measure of turbidity, emergent macrophyte cover and type (i.e., *Bolboschoeno* or *Phragmitetum* dominated), and the shortest distance between the sampled groundwater wells and the shoreline of the corresponding pans are listed in Table 1. The CDOM concentration was almost always higher in the pan (median = 287.6 mg Pt L^{-1}) than in the corresponding groundwater well (median = 112.2 mg Pt L^{-1}; paired t-test logCDOM: $t = 3.912$; $p = 0.002$; Fig 3a). The pH of each pan (median = 9.28) was also significantly higher than the pH of the corresponding well (median = 8.60; paired t-test $t = 3.896$, $p = 0.002$), while the difference between the conductivity of the pans and the corresponding wells was not significant ($median_{pans} = 3.0$ g L^{-1}; $median_{wells} = 1.8$ g L^{-1}; paired t-test $t = 2.086$, $p = 0.057$) (Table 1).

In the pans dominated by *B. maritimus* CDOM was slightly but not significantly higher than in those dominated by *P. australis* ($median_{B.maritimus}$ = 1059 mg Pt L^{-1}; $median_{P.australis}$ = 253 mg Pt L^{-1}; t-test t = 2.269, p = 0.053; Fig. 3c). In the case of the soda pan that completely lacked macrophyte cover the CDOM concentration of the pan was very similar to the CDOM of the groundwater well (Table 1: Unnamed pan). For both the pans dominated by *B. maritimus* and *P. australis* CDOM concentration of the soda pans showed positive relationship to the CDOM concentration of the wells (Fig. 3c), although this correlation was only significant in the case *P. australis* dominated pans (Pearson's *B. maritimus*: r=0.643; p=0.168; *P. australis*: r=0.803; p=0.029). For the pans with *B. maritimus* dominance the non-groundwater related $CDOM_{diff}$ (median=653.5 mg Pt L^{-1}) was also higher (t-test t = 2.603, p = 0.044) than for those dominated by *P. australis* (median=114.94 mg Pt L^{-1}; Fig. 3b).

For the linear model, backward variable selection retained all assessed variables but the water depth and the optical type (Table S2), while the chemical type and the pH of the pans were removed to avoid collinearity (Table S3). The linear model using CDOM and pH of the wells, conductivity of both pans and wells, TSS of then pans, and emergent macrophyte cover and type (Table 1) explained 92.4% of the variation of CDOM content of the pans, and the ANOVA (Fig. 3d, Table S4) revealed that the macrophyte type of the pans significantly explained 45.8% of the variation (p = 0.004), while the CDOM content and the conductivity of the groundwater wells significantly explained 21.2% (p = 0.006) and 13.9% (p = 0.012), respectively. The 9.8% variance explained by the conductivity of the pans, and the 1.2% explained by the extent of the emergent macrophyte cover was not significant (p = 0.052 and p = 0.416, respectively), while the pH of the pans and TSS explained less than 1% and was not significant (p > 0.1).

Experimental release of DOM from macrophytes

After the 29-days-long incubation of the DOM release experiment, the plant material of *P. australis* lost 7.33±0.15% (3.66±0.075 g) dry mass, while *B. maritimus* lost 10.28±2.79% (5.14±1.40 g). The initial CDOM concentration in the experimental bottles was 39.23±2.22 mg Pt L^{-1} and by the end of the experiment it increased in average to 1190 mg Pt L^{-1} and to 3900 mg Pt L^{-1} for *P. australis* and *B. maritimus*, respectively. The total CDOM release from the degraded plant material was 1136 mg Pt g^{-1} dry weight loss and 2675 mg Pt g^{-1} dry weight loss for *P. australis* and *B. maritimus* respectively. According to the Michaelis-Menten kinetics, at the end of the experiment the CDOM concentration was 90.9% and 86.7% of the possible maximum (V_{max}) for *P. australis* and *B. maritimus*, respectively (Fig. 4a).

The initial DOC concentration was 13.40±0.099 mg L^{-1} and by the end of the experiment increased in average to 82 mg L^{-1} and to 183 mg L^{-1} for *P. australis* and *B. maritimus* incubations, respectively. The total DOC released from the degraded plant material was 78 mg g^{-1} dry weight loss and 125 mg g^{-1} dry weight loss for *P. australis* and *B. maritimus*, respectively. According to the Michaelis-Menten kinetics, at the end of the experiment the DOC concentration was 100.9% and 96.8% of the possible maximum (V_{max}) for *P. australis* and *B. maritimus*, respectively (Fig. 4b). The CDOM/DOC ratio increased linearly with significant parameters from 9 to 14.5 and from 16 to 21 for *P. australis* and *B. maritimus*, respectively (Fig. 4c).

Seasonal comparison of a turbid and a colored pan

As expected, the turbid Zab-szék had substantially higher turbidity than the colored Sósér soda pan throughout most of the study period except for two timepoints in summer 2014 (Table 2, Fig 5a). Water depth showed high variation in both pans (Fig 5b), although it was slightly higher in Sósér than in Zab-szék (Table 2). Water depth changes also indicated that the hydrology of the two studied years also followed different patterns as for both pans the beginning of the year in 2014 was characterized by low water depth, which increased from September-October until the end of the year. Meanwhile, in 2017 both pans had higher water levels at the beginning of the year until August, when the water levels decreased and remained low until the end of the study period (Fig 5b). Throughout the two years the pans differed in respect of pH but not conductivity (Table 2, Fig 5c,d). Both CDOM and DOC were higher in Sósér pan than in Zab-szék (Table 2, Fig 5e,f). The concentration of CDOM and DOC also differed between the two years with higher values for both pans in 2014 than in 2017 (Table 2, Fig 5e,f) More precisely, the highest CDOM and DOC levels were all measured in Sósér in 2014 between January and August, which corresponded to the lowest water level period of the given year (Fig. 5b). In this period the mean concentration of CDOM was 6,649 mg Pt L^{-1} and 563 mg L^{-1} for DOC, while for the rest of the year was 1,294 mg Pt L^{-1} and 111 mg L^{-1} for CDOM and DOC, respectively. The two pans also differed in respect of the indicators calculated from the EEMS (Fig 6). Sósér had higher humification index (HIX) throughout the study period, while the freshness index (FRESH) was higher at every timepoint in Zab-szék than in Sósér (Table 2, Fig 6b,c). In addition, both pans had generally higher FRESH values in summer and Zab-szék had a prominent FRESH peak in the beginning of summer (29-May) (Fig 6b). The fluorescence index (FI) did not differ between the two pans (Table 2), although it showed much higher variation in Zab-szék with prominent peaks in summer and autumn (Fig. 6a). For both pans the mean FI index was close to the value expected for DOM of terrestrial and soil origin (Sósér: FI_{median} = 1.235; Zab-szék: FI_{median} = 1.236).

The cross-correlation analyses revealed that both CDOM and DOC were most strongly correlated with TSS, depth, pH and conductivity measured at the same time (lag = 0) except for DOC concentration in Zab-szék in 2014 and its correlation with TSS, depth and pH (Table S5). In the case of Sósér, the lag = 0 correlations between DOC concentration and TSS, depth, pH and conductivity were always significant (positive correlation with TSS, pH and conductivity, and negative with mean)(Table 3). In Zab-szék DOC showed significant (positive) correlation at lag = 0 only with depth and conductivity (Table 3), while for TSS only values measured 2 months earlier (lag = 2) showed significant positive correlation with actual DOC, and for pH only values measured 5 months later (lag = -5) were significantly correlated with actual DOC (Table S5). The CDOM concentration did not significantly correlate with the TSS of the pans, while mean depth, pH and conductivity significantly correlated with CDOM in all cases but pH in Sósér in 2017 and conductivity in Zab-szék in 2014. Interestingly, the sign of the correlations with CDOM depended on the pan: in Sósér CDOM positively correlated with pH and conductivity, and negatively with depth, while in Zab-szék CDOM negatively correlated with pH and conductivity, and positively with depth. In the case of the EEMS indexes significant correlations were only found for DOC in Sósér: FRESH showed significant positive correlation with DOC measured the same time and HIX showed significant negative correlation with DOC measured three months earlier (Table 3, Table S5). In the case of HIX the highest (non-significant) correlations with CDOM in both pans were also detected with CDOM measured three months earlier (Table S5).

Discussion

The results of our study indicate—as we hypothesized—that the CDOM variation in the soda pans of the Carpathian basin is highly influenced by groundwater and the dominant species of the macrophyte cover but—opposite to our hypothesis—it is not significantly driven by the extent of the cover. Furthermore, we demonstrated that the variation of DOC and CDOM concentrations of these pans is related to the variations in pH, conductivity and water depths, and it is presumably influenced by yearly variations in hydroperiods and intrinsic pan properties such as turbidity.

Groundwater as allochthonous source of CDOM

Plant-derived colored DOM in inland waters originates either from macrophytes or from terrestrial vegetation of the watershed via surface or groundwater inflow (i.e., allochthonous source) (V.-Balogh *et al.*, 1998; Wetzel, 2001; Lapierre & Frenette, 2009; Einarsdottir *et al.*, 2017). As no watercourse enters the endorheic intermittent soda pans studied here, the role of surface inflow in their water budget is negligible (Boros *et al.*, 2013), and therefore their plant-derived DOM content is expected to be primarily derived from macrophytes and groundwater inflow. The multisite comparison of polyhumic soda pans demonstrated that the CDOM concentration of pans is related to the CDOM concentration of groundwater (Fig. 3). Groundwaters store and release carbon to surface waters in amounts that are meaningful for global budgets (Downing & Striegl, 2018) and import of terrestrial carbon into lakes via groundwater seepage has been shown to be substantial even in the case of boreal lakes with hydrology dominated by surface water inflow (Einarsdottir *et al.*, 2017). In areas of lower relief, shallow basins can be the focus of local discharge and evaporation from regionally extensive groundwater systems (Deocampo & Jones, 2014) and modern continental evaporates like soda lakes and pans typically accumulate within groundwater discharge (Warren, 2006). Such groundwater driven hydrological processes have been also identified behind the formation of soda pans of the Danube-Tisza Interfluve (Mádl-Szőnyi & Tóth, 2009; Simon *et al.*, 2011). The groundwaters analyzed in this study had relatively high pH (median: 8.60, range: 7.02-9.56, Table 1.), which—considering the pH-dependent dissolution of humic substances—is very likely one of the reasons of their high CDOM content. Considering the high CDOM content of local groundwaters and the importance of groundwater in the hydrology of the soda pans of the region (Simon *et al.*, 2011), it is not surprising that groundwater CDOM explained a substantial part of the variation of the CDOM concentration of the studied soda pans. Finally, the mean FI indexes measured by EEMS in the seasonal monitoring were close to 1.2, which is considered

410 to indicate plant and soil derived organic matter (Fellman *et al.*, 2010) further enforcing the
411 partly terrestrial origin of the DOM of the pans.
412
413 *Autochthonous emergent macrophyte cover type influences CDOM content*
414 However, the relationship between CDOM content of the pans and the corresponding
415 groundwaters depended on the dominant type of emergent macrophytes of the pans with pans
416 dominated by *B. maritimus* having higher CDOM content than those dominated by *P. australis*
417 (Fig. 3). While in the case of lakes macrophyte derived DOC is considered to contribute only
418 to 1-20% of the total DOC, for wetlands and small lakes macrophytes role in the carbon budget
419 can be substantial (Sobek *et al.*, 2006; Reitsema, Meire & Schoelynck, 2018). However, in our
420 linear model macrophyte species dominance was much more important in explaining the
421 variance in CDOM content among the studied pans than the extent of the macrophyte cover
422 (45.8 *vs* 1.2% of variance explained, respectively). A possible explanation to this seemingly
423 surprising result is that in the multisite comparison we focused on CDOM and not total DOC,
424 which depends both on total DOC and the color of DOM. It has been shown that qualitatively
425 macrophyte-derived DOM depends on the type of macrophyte (Qu *et al.*, 2013). Accordingly,
426 it is reasonable that in the given dataset the dominant species of macrophytes is more important
427 in explaining the pans' CDOM content than the extent of emergent macrophyte cover.
428 The importance of the type of vegetation was corroborated by the results of the DOM release
429 experiment, which also showed that more than triple CDOM and more than double the amount
430 of DOC is released from *B. maritimus* than from *P. australis*. While DOM release from *P.*
431 *australis* has been studied before, to the best of our knowledge this is the first report for *B.*
432 *maritimus*. The DOC release results for *P. australis* presented here (82 mg L^{-1}) compare well
433 to those from a previous experiment conducted under similar conditions (75 mg L^{-1}) (V.-Balogh
434 *et al.*, 2006) indicating high reproducibility of our experiment. When compared to other

emergent macrophytes *P. australis* also released less DOC than crofton weed (*Eupatorium adenophorum* (Spreng.) R.M.King & H.Rob., Asteraceae), water oats (*Zizania latifolia* (Griseb.) Turcz., Poaceae), oriental pepper (*Polygonum orientale* (L.) Spach., Polygonaceae) (Qu *et al.*, 2013), or seepweed (*Suaeda salsa* (L.) Pall., Amaranthaceae) (Qi, Xue & Wang, 2017). All these indicates that common reed (*P. australis*) is a relatively low DOC releasing emergent macrophyte and accordingly, it is essential to consider the composition of emergent macrophytes when assessing macrophyte impact on the carbon balance of aquatic systems.

The total release of DOC within the time of the experiment reached values close to the maximum expected for both species (Fig. 4b), while the total CDOM release did not reach the maximum expected for either of the species (Fig. 4a). This contrasting dynamic between CDOM and DOC resulted in continuously increasing ratio of CDOM and DOC (Fig. 4c). To better mimic natural conditions our experiment was not conducted under sterile conditions and oxygen for aerobic microbial degradation was provided by aeration, therefore, the increase in the ratio of CDOM suggest microbial degradation of the labile carbon fraction and concomitant accumulation of recalcitrant colored DOM. The importance of microbial degradation in DOC release was demonstrated by the experiment of Qi, Xue & Wang (2017) were the amount of DOC released from *P. australis* under sterile conditions was nine times higher than the quantity released without inhibition of microbial degradation.

Considering the implications of our experiment to natural conditions, we have to emphasize that the pH of the water used in the experiment was lower (pH 8.40) than the pH of almost all pans analyzed in the multisite comparison (Table 1; pH 9.36). In a decomposition experiment increasing pH has been shown to correlate with increasing dry mass loss from macrophyte litter (Krachler *et al.*, 2010), which together with the pH dependent dissolution of humic substances suggests even higher release of DOC and CDOM from macrophytes in the pans than in our experiment. However, the CDOM/DOC ratio at the end of the experiment was 14 times higher for *P. australis* and 21 times higher for *B. maritimus* than the average ratio of 6.6 measured for 84 soda pans of the Carpathian Basin (Boros *et al.*, 2017). A possible explanation for higher proportion of colorless organic matter in natural habitats is photochemical degradation of CDOM, which has been demonstrated to be a significant contributor to organic carbon mineralization in boreal lakes, and which is expected to be particularly important in shallow high DOC lakes (Koehler *et al.*, 2014). In summary, it is likely that the autochthonous plant-derived DOM of soda pans depends on the species composition of the emergent macrophytes and the interplay of microbial and photochemical degradation.

Potential drivers of the seasonal variation of CDOM and DOC

The seasonal comparison of two soda pans with significant differences in respect of optical characteristics (i.e., colored Sósér and turbid Zab-szék) is reasonable particularly considering the potential importance of photochemical reactions. Sósér and Zab-szék also differ in respect of emergent macrophyte cover (95% and 16%, respectively) and dominant macrophyte species (*B. maritimus* and *P. phragmites*, respectively) (Table 1). Although both pans had much higher CDOM and DOC concentration than those characteristic for other aquatic systems (Fig. 1a), in the case of the colored pan the concentration of both CDOM and DOC was extremely high. Based on the findings of the multisite comparison and DOC release experiment the extensive *B. maritimus* dominated macrophyte cover supposedly contributed to the extreme CDOM and DOC concentrations of Sósér. Meanwhile, compared to Sósér Zab-szék had slightly higher pH, lower water depth and consequently intermittent hydrology with several droughts in 2017. The two pans also differed based on the qualitative EEMS analyses of DOM. The higher HIX index of Sósér suggests higher ratio of plant-derived recalcitrant DOM, while the higher FRESH index of Zab-szék suggests greater ratio of freshly produced autochthonous DOM (Fig 6).

In addition, substantial differences in the hydrology and overall CDOM and DOC concentrations of the two years monitored were also observed. A potential explanation of the contrasting patterns of hydrology of the two years might be in the differences in temperature and precipitation regimes of the study years and the preceding periods. Although weather analyses are beyond the scope of this study, regional records show that between July 2013 and June 2014 the weather was dryer and much warmer than usual. Specifically, the precipitation between July and December of 2013 was only 75% of the monthly mean measured for the region between 1981 and 2010 and in the following period (Jan-Jun 2014) the temperature was 2.3°C higher than the monthly mean (Fig. S1). Meanwhile, for the same period in 2016-2017 both temperature and precipitation were close to average (Fig. S1). Considering the importance of groundwater in the hydrology of the soda pans in this region (Mádl-Szőnyi & Tóth, 2009), the combination of decreased precipitation followed by high temperature in 2013 and 2014 might have caused increased evapotranspiration from soils, leading to decreased groundwater levels, and consequently less groundwater influx to the pans, which explains the lower water levels measured for both pans in the beginning of 2014 compared to 2017. Similarly, the opposite water level trends in autumn (i.e., increase in 2014 and decrease in 2017; Fig. 5b) could be explained with precipitation and temperature anomalies such as the unusually high precipitations in the second half of 2014 (156% of average precipitation; Fig. S1).

In Sósér the lower water level period of 2014 coincided with the highest DOC and CDOM concentrations measured (Fig. 5e,f). In this period the mean DOC concentration of the pan was 563 mg L^{-1}, which is more than an order of magnitude higher than the average of wetlands (Fig. 1a) and remarkable even among soda lakes and pans (e.g. Jirsa *et al.*, 2013; Boros *et al.*, 2017). Both the CDOM and DOC concentrations of Sósér were negatively correlated with water depth throughout the study period (Table 3), which suggests that evaporation driven concentration contributed to the extremely high CDOM and DOC levels measured here. This is supported by the positive correlation of CDOM and DOC to conductivity and pH as both are also expected to increase with decreasing water levels due to the concentration of inorganic ions. Furthermore, considering the pH dependence of the dry matter loss from macrophyte litter (Krachler *et al.*, 2010), the higher pH of the period could have further aggravated the OM release from the dense *B. maritimus* dominated macrophyte cover of Sósér contributing to the measured record high DOC levels. Turbidity positively correlated with DOC suggesting that in this otherwise low turbidity pan (TSS in Zab-szék was almost 20 times higher than in Sósér; Fig. 5a), where organic carbon is the main cause of turbidity opposite to clay minerals, which are typical for turbid type pans such as Zab-szék (Boros *et al.*, 2013).

On the other hand, in Zab-szék the correlation between depth and both CDOM and DOC was positive (Table 3) suggesting that in this groundwater fed pan, possible groundwater itself was the primary source of CDOM and DOC. This explanation was also supported by the results of the multisite comparison, where for this pan—exceptionally among the other studied pans—the CDOM content of the nearby groundwater well was almost double of the CDOM content of the pan (Table 1). Interestingly, in this pan CDOM and DOC correlated differently with pH and conductivity: for CDOM the correlation was negative, while for DOC it was positive. Considering the lower pH and conductivity of the nearby groundwater well, their negative correlations with CDOM agree with its groundwater origin theory. However, the positive correlations of DOC and conductivity are seemingly contradictory to this theory. As DOC measures both labile and recalcitrant organic carbon, while CDOM reflects more recalcitrant organic carbon, a potential explanation to the increasing DOC content at higher conductivity could be the presence of salinity dependent biodegradation inhibition as salinity is one of the strongest microbial inhibitors (Székely et al. 2013). However, it is also possible that the higher DOC but lower CDOM concentrations at high conductivity reflect non-humic freshly produced autochthonous DOM, which together with the lesser macrophyte coverage and the *P.*

phragmites dominance suggests that the influence of phytoplankton and microbial communities on the DOM of this pan could also be prominent. This explanation is enforced by the contrasting microbial community of the two pans described by Szabó *et al.* (2017) and by the peaks of FI index in Zab-szék (Fig 6a), which are potential indicators of phytoplankton and microbial blooms suggesting greater importance of microbial processes in the carbon dynamics of this pan than in Sósér.

In a broader perspective, our results demonstrate that in the case of endorheic water bodies—particularly those lacking surface inflow—groundwater can be an important source of organic carbon that should be accounted for in carbon budget calculations. We also showed that emergent macrophytes are essential sources of recalcitrant organic carbon. Although when estimating macrophyte effect, species composition has to be also considered since common reed (*P. australis*), one of the most common emergent macrophytes on a global scale has relatively low organic carbon release compared to other species such as the cosmopolitan bulrush (*B. maritimus*), for which this study comprises the first report of experimental decomposition measurements. Finally, we demonstrated that the record high DOC values (0.5-1 g L^{-1}) measured in the soda pans of the Carpathian basin are the result of the interplay of intrinsic soda pan characteristics such as *B. maritimus* dominated macrophyte cover and most importantly persistent low water levels that occur in consequence of weather anomalies. More precisely, we showed that high organic carbon content periods follow extreme warm and dry seasons. Considering that such weather patterns might increase in frequency in the near future due to the ongoing climate change, soda pans could become increasingly important hotspots of terrestrial carbon processing urging further studies exploring the carbon biogeochemistry of soda lakes.

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Data Availability Statement

The seasonal monitoring data and fluorescence spectra used for the EEMS analyses are available on the online repository at: http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-385197.

Conflict of Interest Statement

We declare that there is no actual or potential conflict of interest.

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Figure legends

Fig. 1 Global mean (a) and maximum (b) concentrations of dissolved organic carbon (DOC) in inland waters. Data sources: (Wetzel, 2001) for surface water; (Chen *et al.*, 2015) for lakes; (Thurman, 1985) for lakes, swamps, marshes, bogs; Suhett et al (2004) for coastal lagoons; (Boros *et al.*, 2013, 2016, 2017) for soda pans.

Fig. 2 Location of studied soda pans within the Carpathian Basin based on Boros et al., 2017.

Fig. 3 Multisite comparison of the factors related to colored dissolved organic matter (CDOM) in 14 soda pans of the Carpathian Basin. (a) Comparison of CDOM concentration in soda pans and nearby groundwater; (b) Difference between CDOM concentration of soda pans and corresponding groundwater well by dominant macrophyte type of the pans (*Bolboschoenus maritimus* or *Phragmites australis*); (c) Correlation of groundwater and soda pan CDOM. Filled circles indicate *B. maritimus* and crosses *P. australis* dominated pans, while the empty circle indicates no macrophyte cover. Dashed line depicts non-significant ($r = 0.643$; $p = 0.168$) and solid line depicts significant ($r = 0.804$; $p = 0.029$) correlation; (d) Variance of CDOM of the 14 pans explained by the different parameters based on linear model (Table S4). Significance of parameters according to ANOVA indicated by **: $p < 0.01$; *: $p < 0.05$; $p < 0.1$.

Fig. 4 Experimental organic matter release from *Bolboschoenus maritimus* and *Phragmites australis* plant material. (a) Colored dissolved organic matter (CDOM) release. Non-linear Michaelis-Menten kinetics-based curve fitting statistics for *B. maritimus*: N=28; df=25; r^2= 0.7881; V_{max}=4495.6372 p<<0.0001; K_m=5.2482 p=0.0023, and for *P. australis*: N=28; df=26; r^2= 0.8403; V_{max}=1308.8554 p<<0.0001; K_m=5.2816 p=0.0002. (b) Dissolved organic matter (DOC) release. Non-linear Michaelis-Menten kinetics-based curve fitting statistics for *B.*

maritimus: N=28; df=26; r^2= 0.9197; V_{max}= 189.4330 $p<<0.0001$; K_m= 1.9014 $p<<0.0001$, and for *P. australis*: N=28; df=26; r^2= 0.8596; V_{max}= 81.2560 $p<<0.0001$; K_m= 1.1827 $p<<0.0001$. (c) Changes CDOM/DOC. Linear curve fitting statistics for *B. maritimus*: N=28; df=26; r^2= 0.5630; intercept= 15.7668 $p<<0.0001$; slope= 0.2204 p=0.0028 and for *P. australis*: N=28; df=26; r^2=0.7529; intercept= 9.3597 $p<<0.0001$; slope= 0.1896 $p<<0.0001$.

Fig. 5 Multiyear comparison of the temporal changes of environmental parameters in Sósér and Zab-szék soda pans in 2014 and 2017. (a) Total suspended solids (TSS); (b) Mean water depth; (c) pH; (d) Electrical conductivity (EC); (e) Colored dissolved organic matter (CDOM); (f) dissolved organic carbon (DOC).

Fig. 6 Temporal changes of dissolved organic matter (DOM) characterizing indices derived from fluorescence excitation emission matrix spectroscopy (EEMS) in Sósér and Zab-szék soda pans in 2017. (a) Fluorescence index (FI) indicating the source of DOM with high values (FI $\approx$ 1.8) suggesting DOM derived from extracellular release and leachate from bacteria and algae, and low values (FI $\approx$ 1.2) indicating plant and soil derived organic matter. (b) Freshness index (FRESH) indicating contribution of recently produced DOM. (c) Humification index (HIX) indicating humic substance content or extent of humification of DOM (Fellman *et al.*, 2010).

Table 1. Environmental parameters determined for the soda pans and corresponding groundwater wells in the multisite comparison

	Soda pan									Groundwater well			
Name of pan	Chemical type*	Optical type†	Emergent macrophyte cover (%)	Macrophyte type (species cover > 50%)	TSS (mg L^{-1})	Electrical conductivity ($\mu S\ cm^{-1}$)	pH	CDOM (Pt mg L^{-1})	Water depth (cm)	Electrical conductivity ($\mu S\ cm^{-1}$)	pH	CDOM (Pt mg L^{-1})	Distance to closest shore of soda pan (m)
Bogárzó	Soda	Turbid	43	*B. maritimus*	807	4970	9.28	1424	30	5460	8.55	107	34
Böddi-szék 1	Soda-Saline	Turbid	15	*P. australis*	745	6420	9.80	239	9	2810	8.83	74	24
Böddi-szék 2	Soda-Saline	Turbid	5	*P. australis*	1100	1710	9.69	253	2	5100	9.56	117	16
Böddi-szék 3	Soda-Saline	Colored	92	*P. australis*	8	1847	8.39	293	40	3400	9.29	303	239
Büdös-szék	Soda	Turbid	62	*B. maritimus*	384	5230	9.27	174	12	2200	7.73	28	190
Csaba-szék	Soda	Colored	52	*B. maritimus*	50	2620	8.57	1659	20	1863	8.34	841	25
Dongér	Soda-Saline	Turbid	11	*P. australis*	67	1625	10.57	99	20	757	8.64	67	31
Fehér-szék	Soda	Turbid	91	*P. australis*	585	3300	8.86	684	45	2310	8.74	437	155
Fülöp-szék	Soda	Colored	75	*P. australis*	350	4090	9.12	143	8	4010	7.02	28	198
Kelemen-szék	Soda	Turbid	30	*B. maritimus*	1850	3010	9.13	693	20	1146	8.15	205	201
Sósér	Soda	Colored	95	*B. maritimus*	83	8680	9.89	2088	3	3540	9.28	69	47
Unnamed	Soda	Turbid	0	none	692	4840	9.43	203	20	1609	8.73	146	85
Vesszős-szék	Soda	Turbid	72	*B. maritimus*	80	2050	9.73	282	20	1678	8.17	41	467
Zab-szék	Soda	Turbid	16	*P. australis*	1574	4270	9.28	364	3	1051	8.30	667	45

* Based on Boros & Kolpakova (2018)

† Based on Boros *et al.*, (2013, 2017)

Table 2. Mean and standard deviation of the measured parameters of the turbid and colored soda pan in the two years of seasonal monitoring

Parameter	Sósér				Zab-szék			
	2014		2017		2014		2017	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
TSS	180.8	293.0	68.7	71.1	2325.9	2201.9	2100.9	1618.3
Depth	18.6	15.3	26.8	14.3	13.1	12.1	11.3	9.6
pH	9.26	0.54	9.23	0.40	9.60	0.28	9.61	0.21
EC	5.72	3.45	6.49	3.79	5.64	3.66	7.05	6.68
CDOM	5888.7	2908.6	1628.2	508.7	470.4	200.8	279.1	127.3
DOC	437.3	268.5	214.6	108.4	69.1	48.0	49.4	10.8
FI	-	-	1.228	0.028	-	-	1.234	0.049
FRESH	-	-	0.608	0.030	-	-	0.736	0.091
HIX	-	-	17.47	1.689	-	-	7.143	1.436

Table 3. Results of cross-correlation at lag = 0 of colored dissolved organic matter (CDOM) and dissolved organic carbon (DOC) and environmental parameters of the turbid and colored soda pans. Significant correlations ($p < 0.05$) are in bold.

	CDOM			DOC		
	Sósér		Zab-szék*	Sósér		Zab-szék*
	2014	2017	2014	2014	2017	2014
TSS	0.316	0.598	0.355	**0.737**	**0.745**	0.122
Depth	**-0.750**	**-0.829**	**0.809**	**-0.758**	**-0.778**	**0.622**
pH	**0.778**	0.563	**-0.792**	**0.930**	**0.839**	0.414
Conductivity	**0.802**	**0.660**	-0.447	**0.994**	**0.870**	**0.951**
FI		-0.297			-0.568	
FRESH		0.181			**0.809**	
HIX		-0.225			-0.125	

* Measurements from 2017 for Zab-szék were omitted from these analyses because of missing data due to drought of the pan.

Figure 1

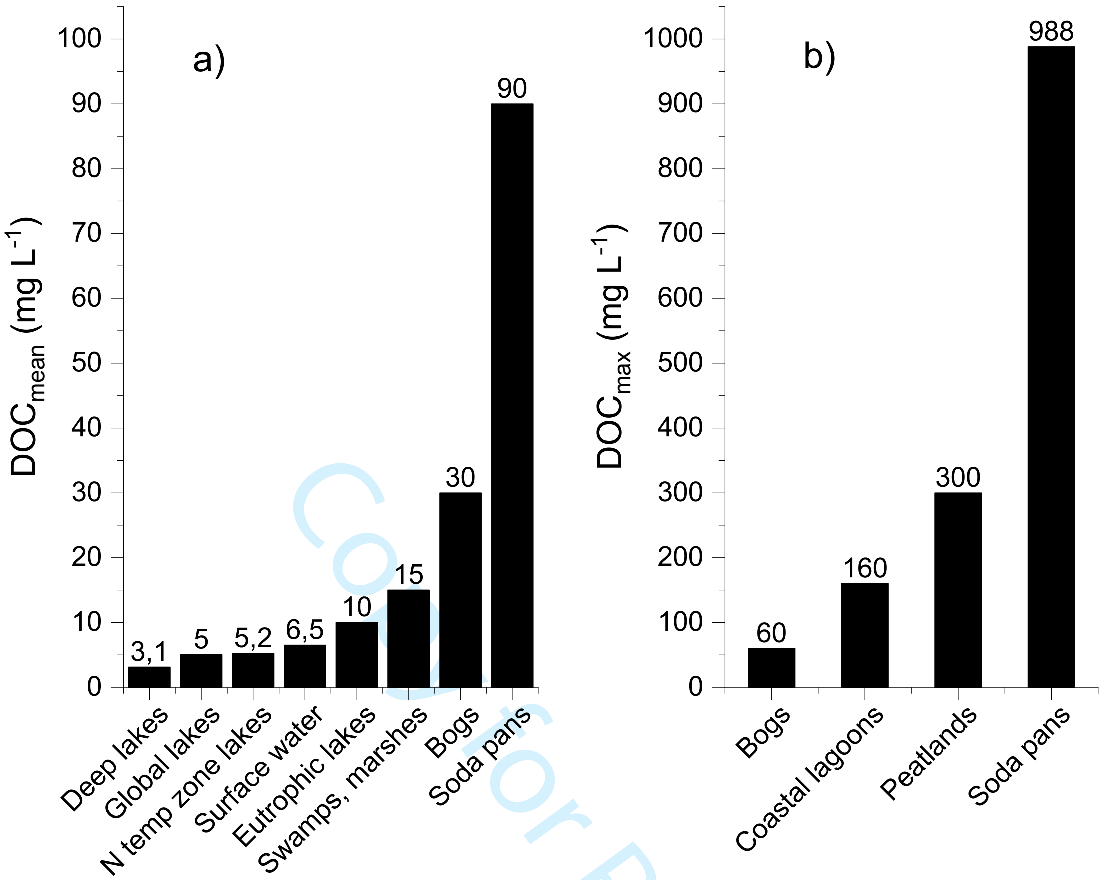

a)
DOC$_{mean}$ (mg L^{-1})
0
10
20
30
40
50
60
70
80
90
100
3,1
5
5,2
6,5
10
15
30
90
Deep lakes
Global lakes
N temp zone lakes
Surface water
Eutrophic lakes
Swamps, marshes
Bogs
Soda pans
b)
DOC$_{max}$ (mg L^{-1})
0
100
200
300
400
500
600
700
800
900
1000
60
160
300
988
Bogs
Coastal lagoons
Peatlands
Soda pans

Figure 2

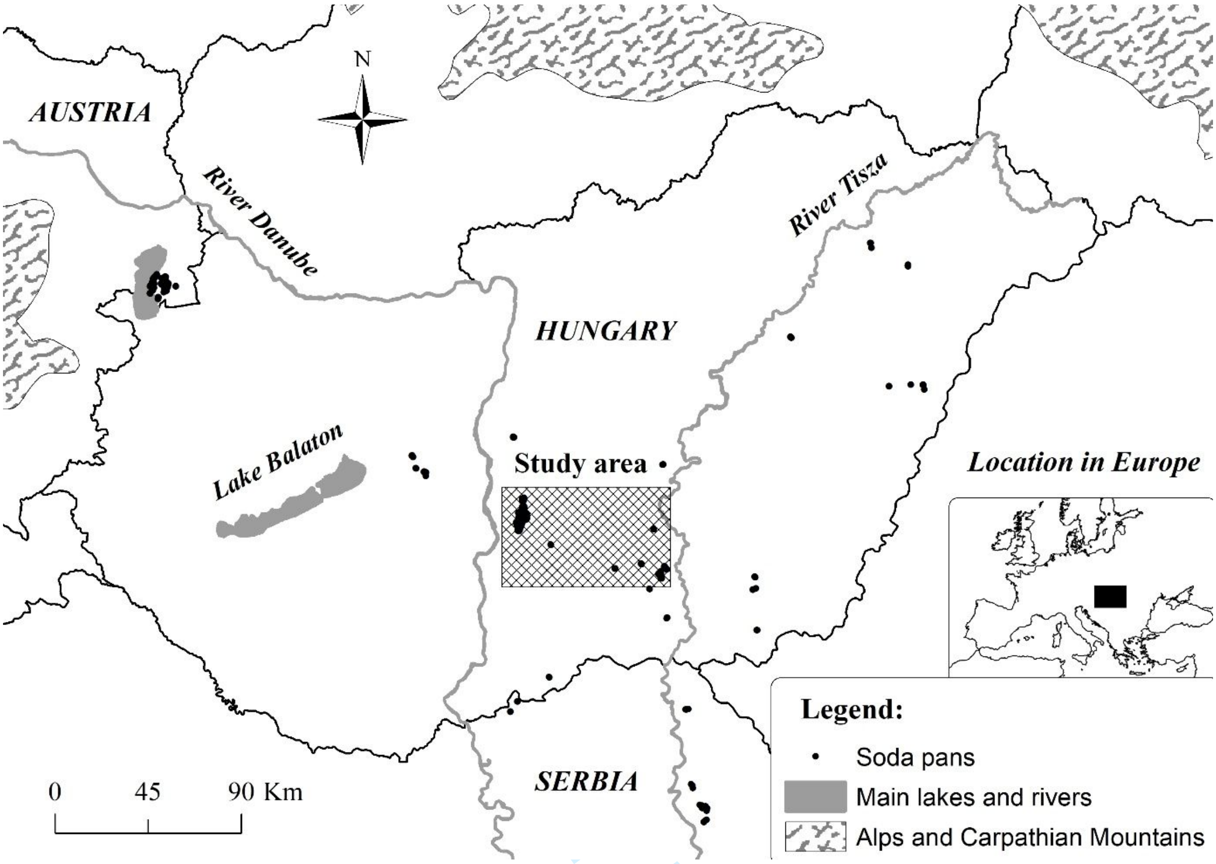

AUSTRIA
N
River Danube
River Tisza
HUNGARY
Lake Balaton
Study area
Location in Europe
SERBIA
0 45 90 Km
Legend:
Soda pans
Main lakes and rivers
Alps and Carpathian Mountains

Figure 3

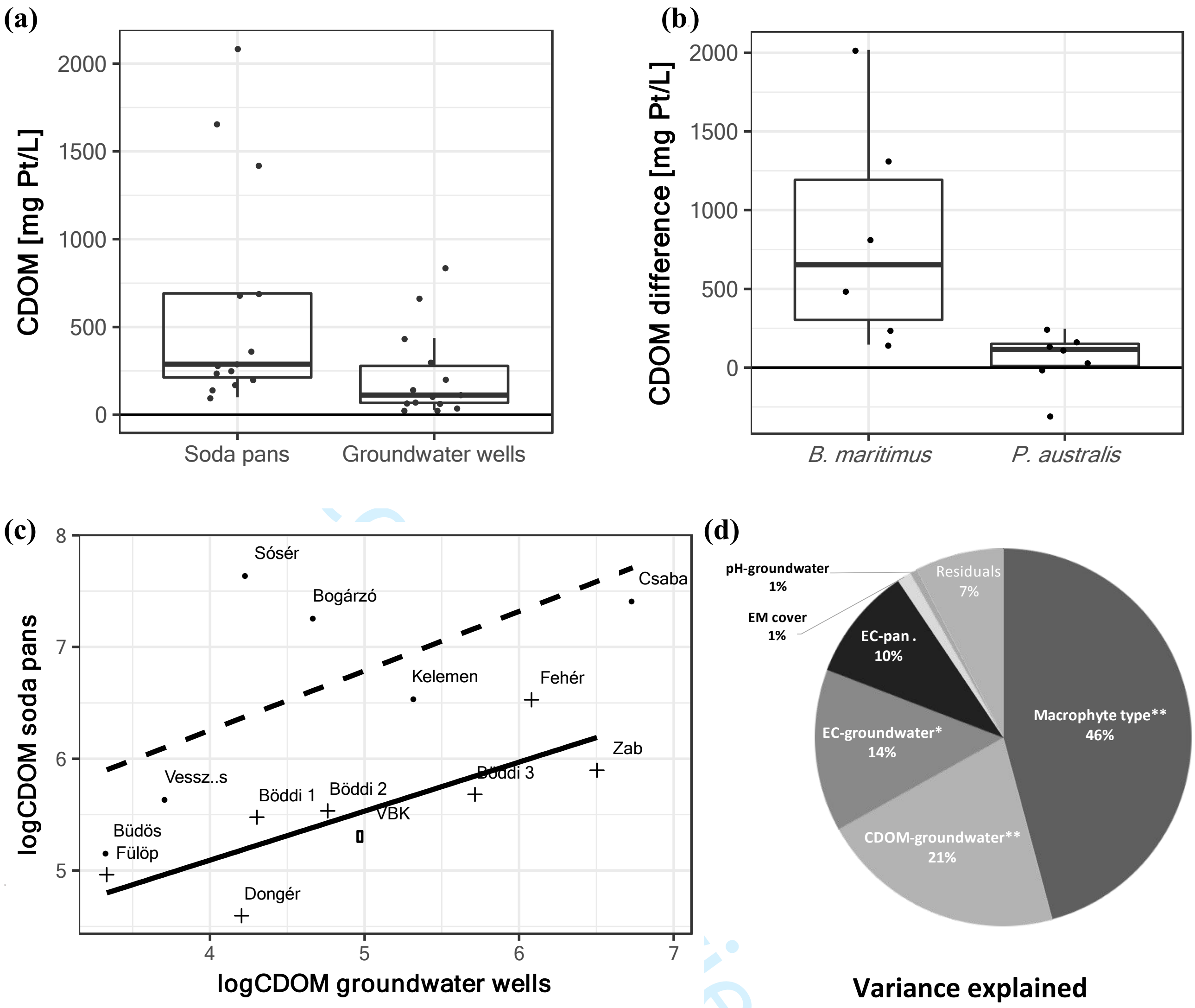
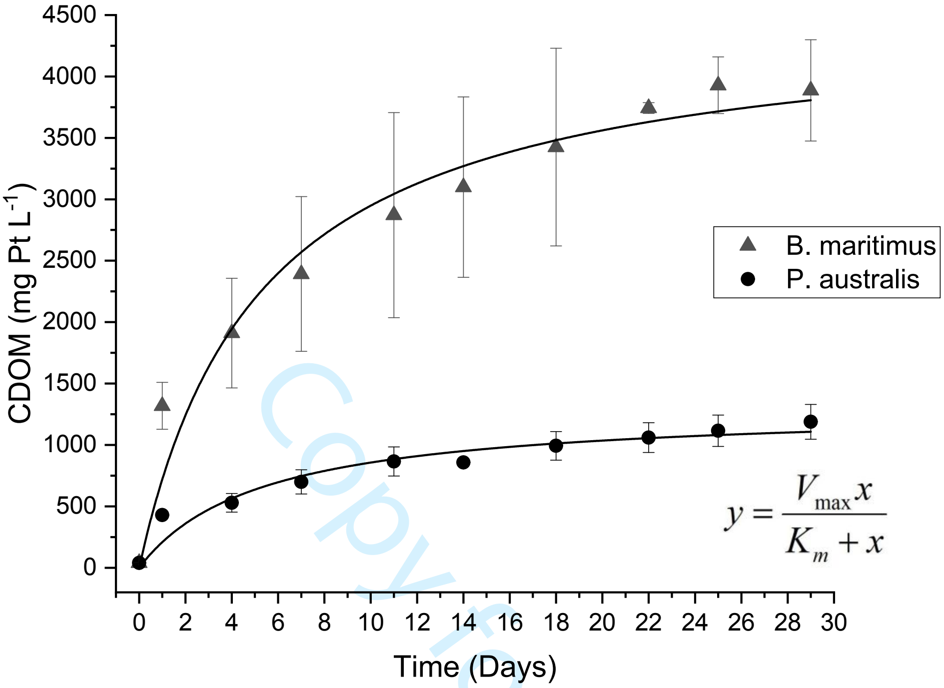
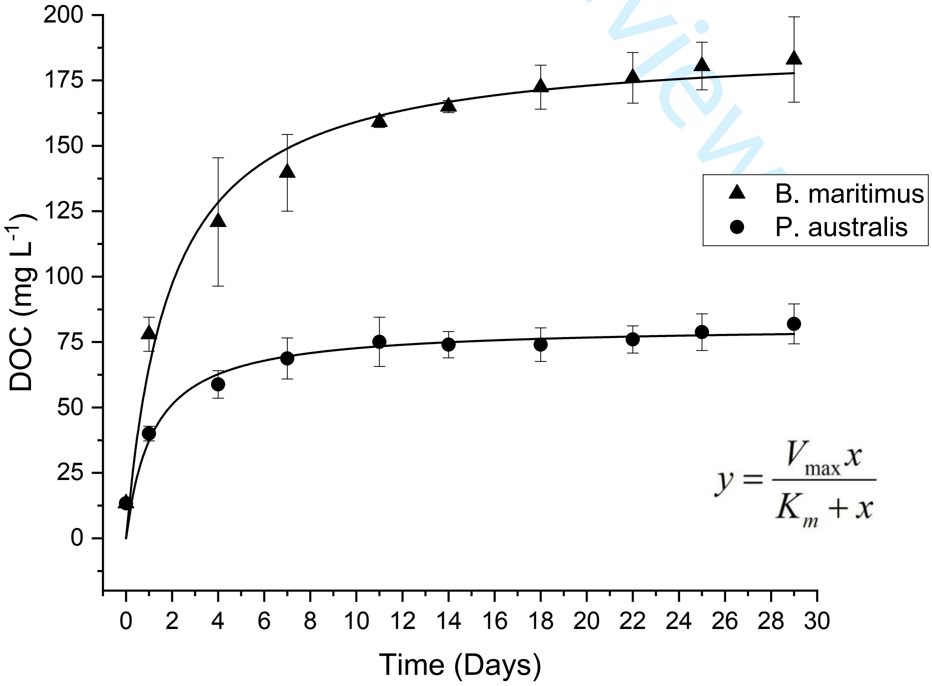

(a)
CDOM [mg Pt/L]
2000
1500
1000
500
0
Soda pans
Groundwater wells
(b)
CDOM difference [mg Pt/L]
2000
1500
1000
500
0
B. maritimus
P. australis
(c)
logCDOM soda pans
8
7
6
5
Sósér
Bogárzó
Csaba
Kelemen
Fehér
Zab
Vessz..s
Böddi 1
Böddi 2
Böddi 3
VBK
Büdös
Fülöp
Dongér
4
5
6
7
logCDOM groundwater wells
(d)
pH-groundwater 1%
EM cover 1%
Residuals 7%
EC-pan . 10%
Macrophyte type** 46%
EC-groundwater* 14%
CDOM-groundwater** 21%
Variance explained

Figure 4

(a)


CDOM (mg Pt L-1)
Time (Days)
B. maritimus
P. australis
$y = \frac{V_{max} x}{K_m + x}$

(b)


DOC (mg L-1)
Time (Days)
B. maritimus
P. australis
$y = \frac{V_{max} x}{K_m + x}$

(c)

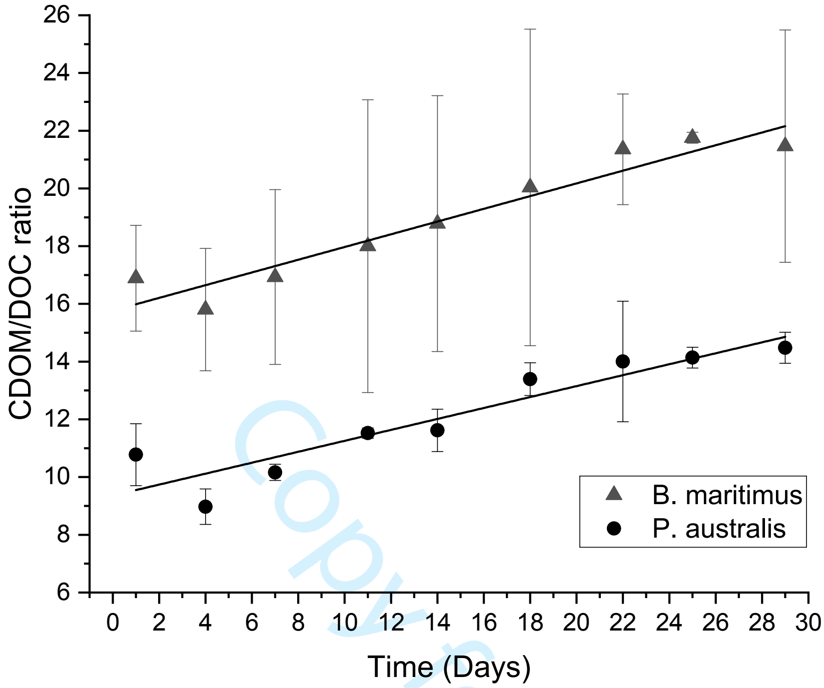

CDOM/DOC ratio
Time (Days)
B. maritimus
P. australis
26
24
22
20
18
16
14
12
10
8
6
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30

Figure 5

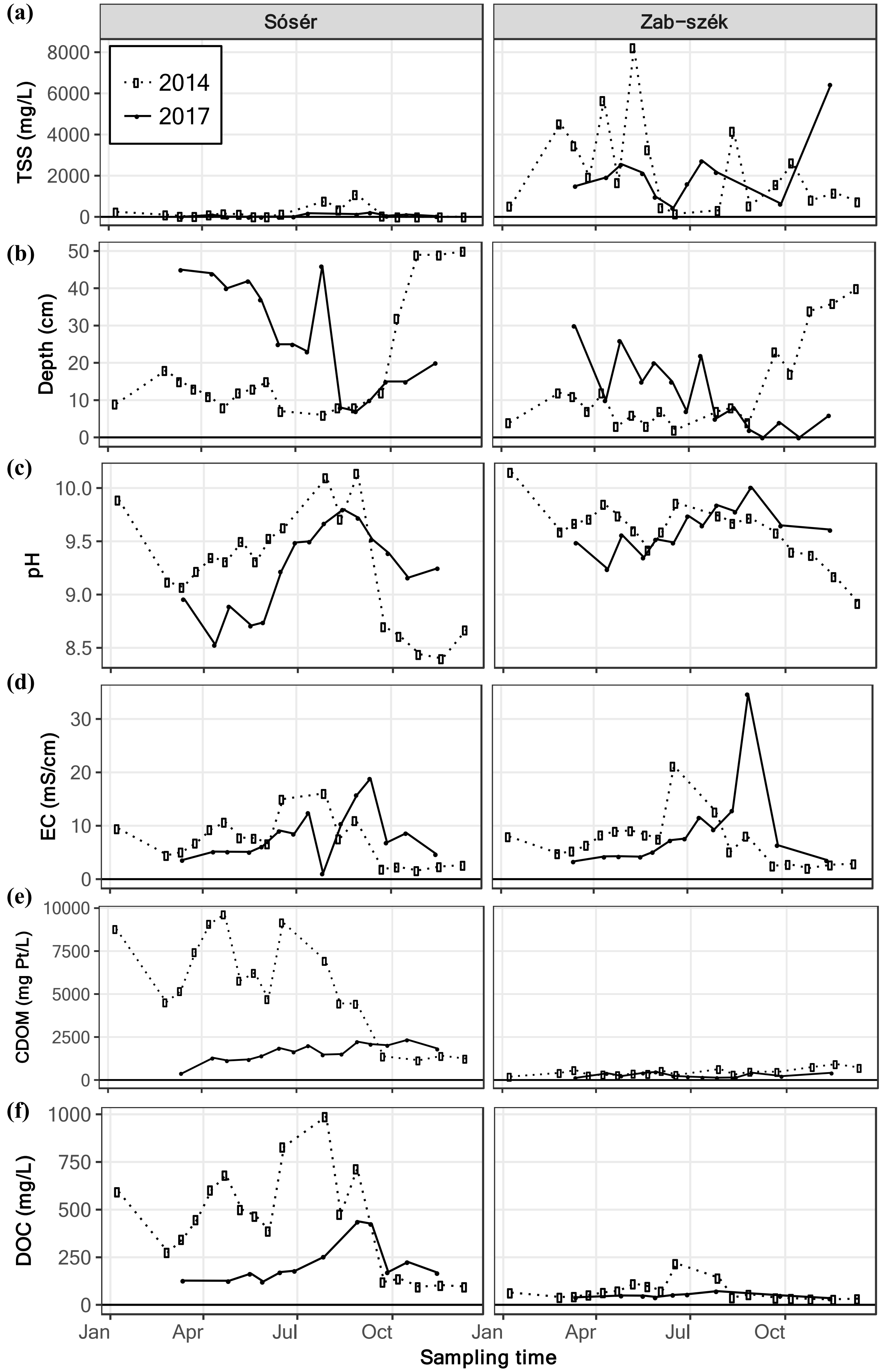
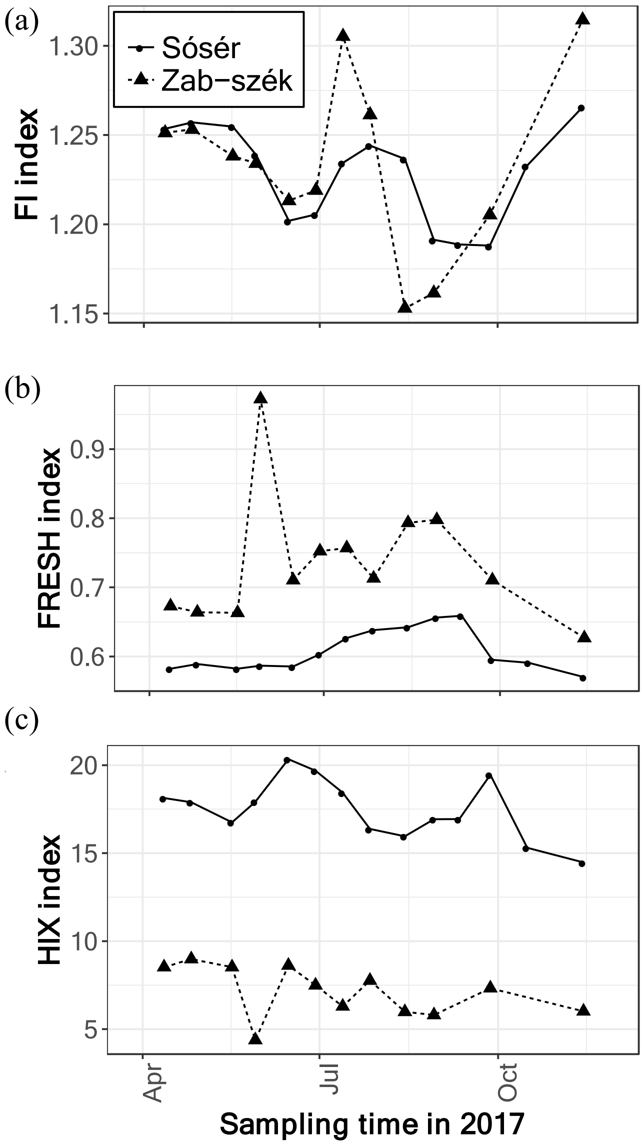

(a)
Sósér
Zab−szék
2014
2017
TSS (mg/L)
0
2000
4000
6000
8000
(b)
Depth (cm)
0
10
20
30
40
50
(c)
pH
8.5
9.0
9.5
10.0
(d)
EC (mS/cm)
0
10
20
30
(e)
CDOM (mg Pt/L)
0
2500
5000
7500
10000
(f)
DOC (mg/L)
0
250
500
750
1000
Jan
Apr
Jul
Oct
Sampling time

Figure 6



(a)
Sósér
Zab−szék
FI index
1.30
1.25
1.20
1.15
(b)
FRESH index
0.9
0.8
0.7
0.6
(c)
HIX index
20
15
10
5
Apr
Jul
Oct
Sampling time in 2017

Table S1. Coordinates of soda pans and groundwater wells studied I the cross-sectional analyses.

	Soda pans		Groundwater wells	
Name of pan	Latitude (N)	Longitude (E)	Latitude (N)	Longitude (E)
Bogárzó	46.8054	19.1412	46.8045	19.1403
Böddi-szék 1	46.7666	19.1500	46.7688	19.1500
Böddi-szék 2	46.7603	19.1476	46.7604	19.1463
Böddi-szék 3	46.7692	19.1229	46.7684	19.1246
Büdös-szék	46.5467	20.0298	46.5489	20.0259
Csaba-szék	46.8182	19.1888	46.8179	19.1875
Dongér	46.5725	20.0593	46.5725	20.0583
Fehér-szék	46.8083	19.1867	46.8106	19.1845
Fülöp-szék	46.5787	19.9877	46.5775	19.9889
Kelemen-szék	46.7973	19.1743	46.8012	19.1717
Sósér	46.7877	19.1350	46.7866	19.1355
Unknown	46.7636	19.1807	46.7642	19.1807
Vesszős-szék	46.5249	20.0373	46.5249	20.0310
Zab-szék	46.8342	19.1748	46.8352	19.1747

Table S2. Summary of backward selection of parameter for AIC minimalization

Initial model:
log(CDOM$_{\text{soda pans}}$) ~ log(CDOM$_{\text{groundwater wells}}$) + Conductivity$_{\text{soda pans}}$ + Conductivity$_{\text{groundwater wells}}$ + pH$_{\text{soda pans}}$ + pH$_{\text{groundwater wells}}$ + Emerged macrophyte type + Emerged macrophyte cover + Depth + TSS + Chemical type + Optical type

Model after backward selection:
log(CDOM$_{\text{soda pans}}$) ~ log(CDOM$_{\text{groundwater wells}}$) + Conductivity$_{\text{soda pans}}$ + Conductivity$_{\text{groundwater wells}}$ + pH$_{\text{soda pans}}$ + pH$_{\text{groundwater wells}}$ + Emerged macrophyte type + Emerged macrophyte cover + TSS + Chemical type

Step	Removed parameter	Df	Deviance	Residuals Df	Residuals Deviance	AIC
1				1	0.2145496	-32.49581
2	Depth	1	0.00065744	2	0.2152070	-34.45297
3	Optical type	1	0.00077518	3	0.2159822	-36.40263

Table S3. Summary of GVIF (Generalized Variance Inflation Factors) analyses before and after removal of inflated parameters (GVIF^(1/(2*Df))^2 > 5). Analyses were performed using *vif* function of *car* package in R.

Final model after backward selection:
log($CDOM_{soda\ pans}$) ~ log($CDOM_{groundwater\ wells}$) + $Conductivity_{soda\ pans}$ + $Conductivity_{groundwater\ wells}$ + $pH_{soda\ pans}$ + $pH_{groundwater\ wells}$ + Emerged macrophyte type + Emerged macrophyte cover + TSS + Chemical type

	GVIF	**Df**	**GVIF^(1/(2*Df))**
log($CDOM_{groundwater\ wells}$)	21.948539	1	4.684927
$Conductivity_{groundwater\ wells}$	4.796074	1	2.189994
$Conductivity_{soda\ pans}$	1.581812	1	1.257701
$pH_{groundwater\ wells}$	20.822963	1	4.563218
$pH_{soda\ pans}$	18.047921	1	4.248285
Emerged macrophyte type	20.992869	2	2.140513
Emerged macrophyte cover	21.216754	1	4.606165
TSS	5.424868	1	2.329135
Chemical type	24.508659	1	4.950622

Model after removal of highest GVIF parameter (Chemical type):
log($CDOM_{soda\ pans}$) ~ log($CDOM_{groundwater\ wells}$) + $Conductivity_{soda\ pans}$ + $Conductivity_{groundwater\ wells}$ + $pH_{soda\ pans}$ + $pH_{groundwater\ wells}$ + Emerged macrophyte type + Emerged macrophyte cover + TSS

	GVIF	**Df**	**GVIF^(1/(2*Df))**
log($CDOM_{groundwater\ wells}$)	4.707523	1	2.169683
$Conductivity_{groundwater\ wells}$	2.288839	1	1.512891
$Conductivity_{soda\ pans}$	1.551452	1	1.245573
$pH_{groundwater\ wells}$	2.555346	1	1.598545
$pH_{soda\ pans}$	5.345451	1	2.312023
Emerged macrophyte type	2.216132	2	1.22011
Emerged macrophyte cover	4.035098	1	2.008755
TSS	2.057321	1	1.434337

Final model after removal of highest GVIF parameter ($pH_{soda\ pans}$):
log($CDOM_{soda\ pans}$) ~ log($CDOM_{groundwater\ wells}$) + $Conductivity_{soda\ pans}$ + $Conductivity_{groundwater\ wells}$ + $pH_{soda\ pans}$ + $pH_{groundwater\ wells}$ + Emerged macrophyte type + Emerged macrophyte cover + TSS

	GVIF	**Df**	**GVIF^(1/(2*Df))**
log($CDOM_{groundwater\ wells}$)	1.740759	1	1.319378
$Conductivity_{groundwater\ wells}$	1.41167	1	1.188137
$Conductivity_{soda\ pans}$	1.337339	1	1.156434
$pH_{groundwater\ wells}$	1.533761	1	1.238451
Emerged macrophyte type	1.615174	2	1.12734

Emerged macrophyte cover	2.270607	1	1.506853
TSS	1.955604	1	1.398429

Table S4. ANOVA table of the final linear model from table S3 ordered by variance explained. Significant correlations (p < 0.05) are in bold. Significance codes: $p < 0.001$: '***'; $p < 0.01$: '**'; $p < 0.05$:'*'; $p < 0.1$ '.'; $p > 0.1$ ' '

	Df	**Sum Sq**	**Mean Sq**	**F value**	**Pr (>F)**	**Variance explained (%)**	**Significance code**
Emerged macrophyte type	2	5.409892	2.704946	15.099960	0.007605	45.783090	**
log(CDOM$_{groundwater\ wells}$)	1	2.499060	2.499060	13.950630	0.013500	21.149160	**
Conductivity$_{groundwater\ wells}$	1	1.646254	1.646254	9.189968	0.029032	13.932000	*
Conductivity$_{soda\ pans}$	1	1.152705	1.152705	6.434803	0.052103	9.755164	.
Emerged macrophyte cover	1	0.140855	0.140855	0.786304	0.415828	1.192038	
pH$_{groundwater\ wells}$	1	0.071898	0.071898	0.401362	0.554230	0.608466	
TSS	1	0.000009	0.000009	0.000051	0.994592	0.000077	
Residuals	5	0.895680	0.179136	NA	NA	7.580003	

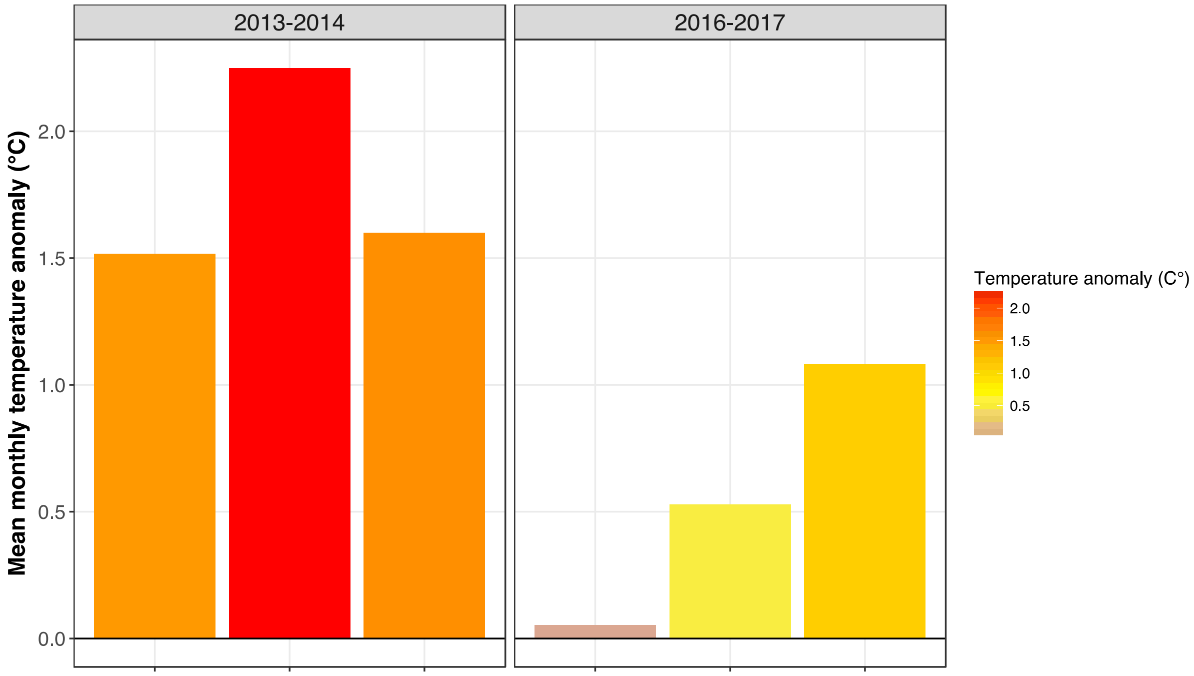
Table S5. Results of cross-correlation analyses between CDOM and DOC concentrations and TSS, mean depth, pH, EC, and EEMS indexes (FI, FRESH and HIX) according to $x_{t+k} \sim y_t$, where x_{t+k} is CDOM or DOM at timepoint $t+k$, and y_t is the tested parameter at timepoint t, $k = 1$ corresponds to 1 month lag. Significant correlations ($p < 0.05$) are in bold. Only correlations between k = -5 and k = 5 are presented as no greater lag cross-correlations were significant.

TSS								**k**					
Variable	**Pan**	**Year**	-5	-4	-3	-2	-1	0	1	2	3	4	5
CDOM	Sósér	2014	-0.159	0.227	0.331	0.181	0.37	0.316	-0.188	-0.412	-0.378	-0.4	-0.126
CDOM	Sósér	2017	-0.376	-0.566	0.049	0.265	0.314	0.598	0.407	0.323	0.108	-0.218	-0.296
CDOM	Zab-szék	2014	0.182	-0.183	-0.049	-0.097	-0.479	-0.355	-0.311	-0.016	-0.156	-0.333	0.045
DOC	Sósér	2014	-0.346	-0.087	0.082	0.113	**0.63**	**0.737**	-0.033	-0.426	-0.405	-0.473	-0.197
DOC	Sósér	2017	-0.219	-0.416	-0.356	0.143	0.482	**0.745**	**0.666**	-0.058	-0.363	-0.343	-0.282
DOC	Zab-szék	2014	-0.187	-0.031	-0.091	-0.147	-0.151	-0.122	0.371	**0.702**	0.374	0.195	0.008
Depth													
CDOM	Sósér	2014	0.115	-0.035	-0.404	**-0.645**	**-0.735**	**-0.75**	-0.315	-0.033	0.132	0.234	0.225
CDOM	Sósér	2017	0.499	-0.001	-0.006	-0.438	-0.603	**-0.829**	-0.466	-0.248	0.137	0.194	0.321
CDOM	Zab-szék	2014	0.106	0.162	0.156	0.44	**0.663**	**0.809**	0.495	0.2	-0.025	-0.198	-0.245
DOC	Sósér	2014	0.447	0.446	0.037	-0.453	**-0.718**	**-0.758**	-0.377	-0.093	0.043	0.134	0.165
DOC	Sósér	2017	0.37	0.209	-0.039	-0.39	**-0.669**	**-0.778**	-0.246	-0.066	0.305	0.411	0.482
DOC	Zab-szék	2014	**0.738**	0.449	0.068	-0.287	**-0.598**	**-0.622**	-0.429	-0.227	-0.06	0.048	0.041
pH													
CDOM	Sósér	2014	-0.212	0.072	0.277	0.425	**0.626**	**0.778**	0.267	-0.024	-0.113	-0.376	-0.22
CDOM	Sósér	2017	-0.437	-0.44	-0.264	0.324	0.57	0.563	0.418	0.349	0.126	-0.124	-0.372
CDOM	Zab-szék	2014	-0.133	0	-0.116	-0.34	-0.564	**-0.792**	-0.358	-0.165	-0.001	0.065	0.014
DOC	Sósér	2014	-0.403	-0.35	-0.145	0.124	**0.646**	**0.93**	0.406	0.035	-0.113	-0.371	-0.237
DOC	Sósér	2017	-0.226	-0.324	-0.304	-0.066	**0.444**	**0.839**	0.726	0.254	-0.431	-0.598	-0.365
DOC	Zab-szék	2014	**-0.664**	-0.275	0.111	0.339	0.441	0.414	0.096	0.009	0.103	0.099	0.201

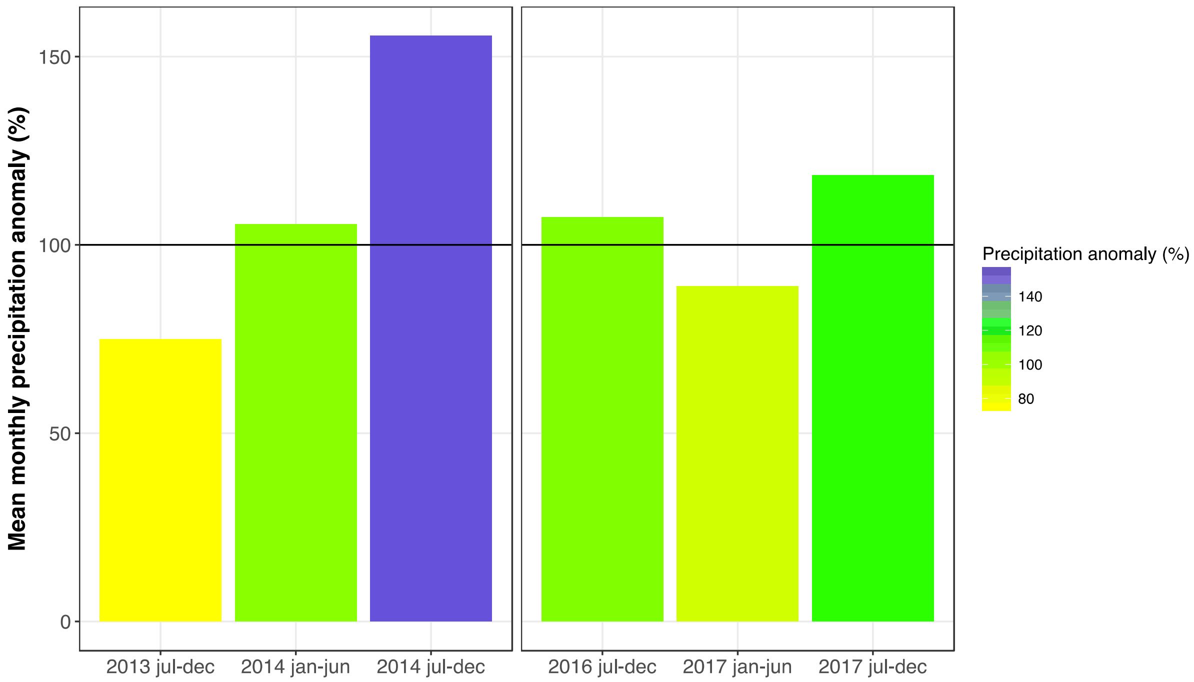
EC								**k**					
Variable	**Pan**	**Year**	**-5**	**-4**	**-3**	**-2**	**-1**	**0**	**1**	**2**	**3**	**4**	**5**
CDOM	Sósér	2014	-0.209	0.017	0.394	0.36	0.535	**0.802**	0.304	-0.051	-0.171	-0.432	-0.41
CDOM	Sósér	2017	-0.593	-0.179	-0.161	0.173	0.236	**0.66**	0.529	0.349	-0.032	-0.096	-0.22
CDOM	Zab-szék	2014	-0.179	-0.108	-0.246	-0.369	-0.384	-0.447	-0.377	-0.338	0.061	0.444	0.502
DOC	Sósér	2014	-0.39	-0.337	-0.053	0.015	0.469	**0.994**	0.542	0.059	-0.066	-0.389	-0.464
DOC	Sósér	2017	-0.322	-0.334	-0.448	0.027	0.627	**0.87**	0.326	0.104	-0.199	-0.291	-0.42
DOC	Zab-szék	2014	-0.515	-0.563	-0.349	0.093	**0.673**	**0.951**	**0.62**	0.125	-0.164	-0.401	-0.344
FI													
CDOM	Sósér	2017	0.344	0.114	0.314	-0.029	-0.115	-0.297	-0.413	-0.175	-0.017	-0.04	0.158
CDOM	Zab-szék	2017	0.143	-0.598	-0.143	0.296	0.028	0.207	0.303	-0.211	-0.344	0.188	-0.104
DOC	Sósér	2017	0.176	0.235	0.578	0.132	-0.538	-0.568	-0.175	-0.197	0.163	0.32	0.181
FRESH													
CDOM	Sósér	2017	-0.508	-0.513	-0.159	0.079	0.144	0.181	0.377	0.373	0.159	-0.132	-0.218
CDOM	Zab-szék	2017	-0.092	0.208	0.207	-0.169	0.081	-0.02	-0.468	-0.225	0.387	-0.077	0.071
DOC	Sósér	2017	-0.164	-0.396	-0.617	-0.275	0.365	**0.89**	0.624	-0.027	-0.387	-0.356	-0.059
HIX													
CDOM	Sósér	2017	0.079	-0.123	-0.529	-0.389	-0.575	-0.225	0.038	0.103	0.21	0.349	0.16
CDOM	Zab-szék	2017	0.284	0.155	-0.44	0.244	0.067	-0.362	0.4	0.042	-0.578	0.058	0.09
DOC	Sósér	2017	0.153	-0.039	**-0.661**	-0.609	-0.201	-0.125	0.103	0.454	0.274	0.173	-0.055

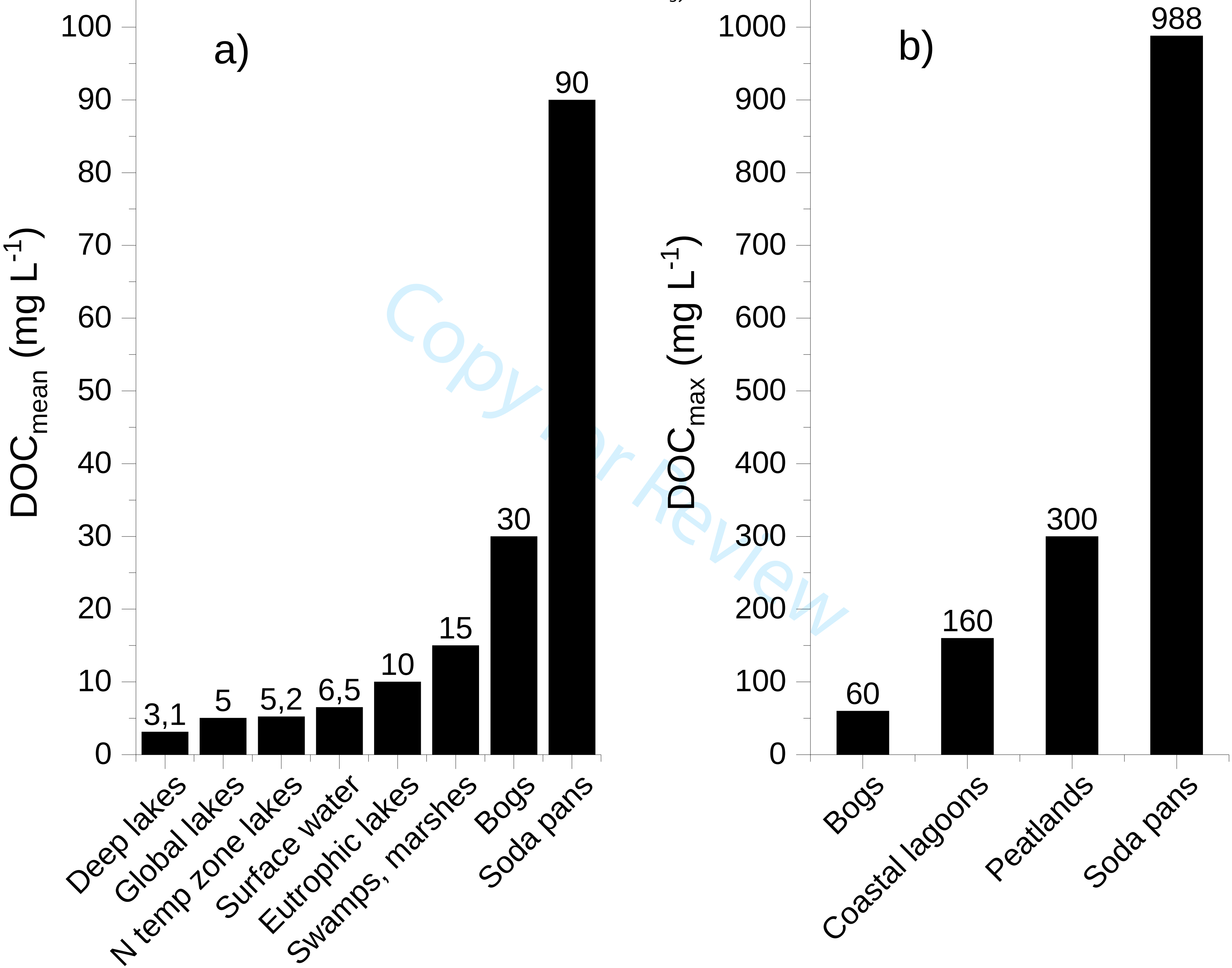
Fig. S1. Temperature and precipitation anomalies before and during the sampling periods of the seasonal analysis as six-months mean of difference from mean monthly average measured between 1981-2010. (a) Temperature and (b) precipitation. Based on data from Hungarian Meteorological Service (OMSz).

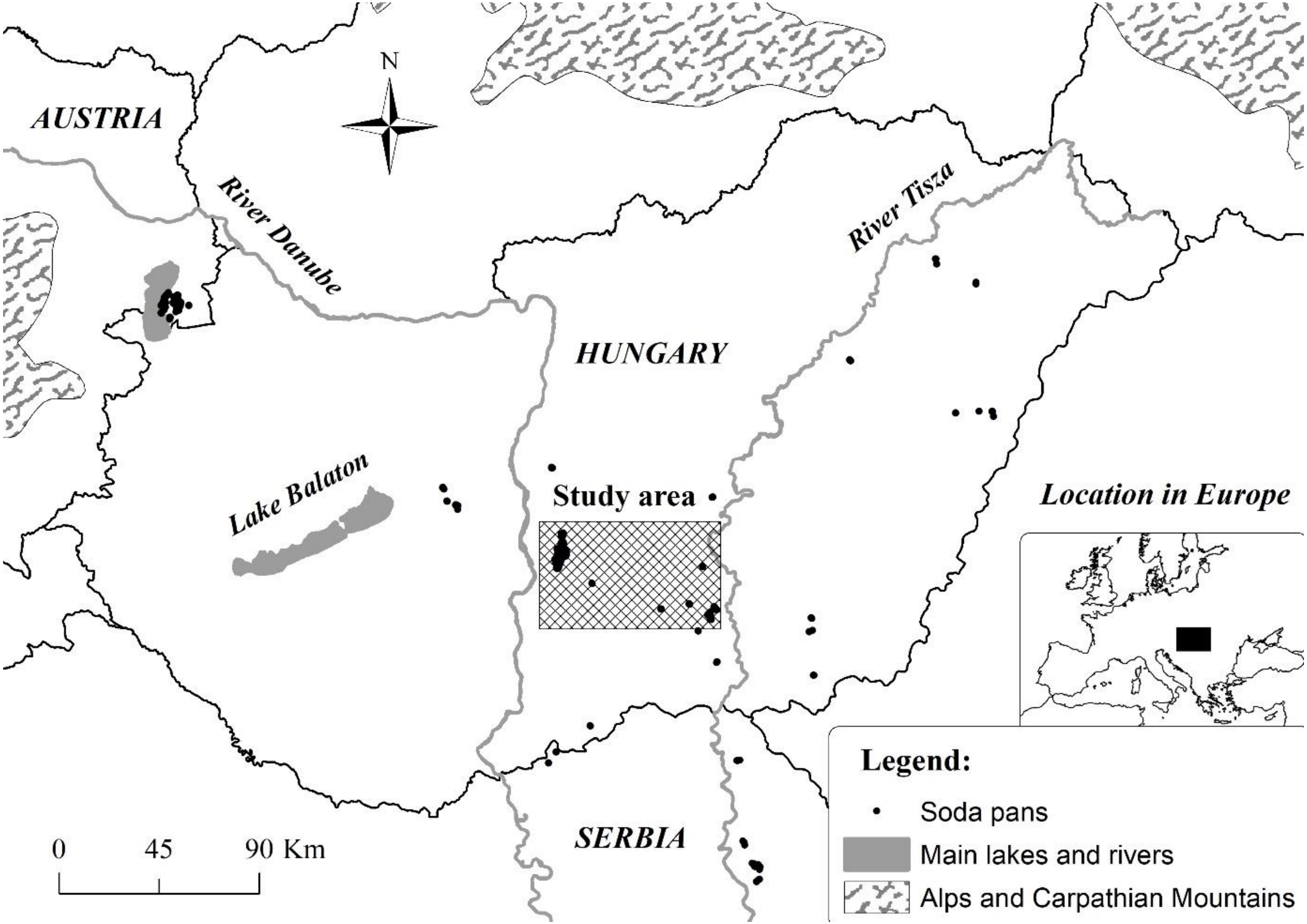
(a)

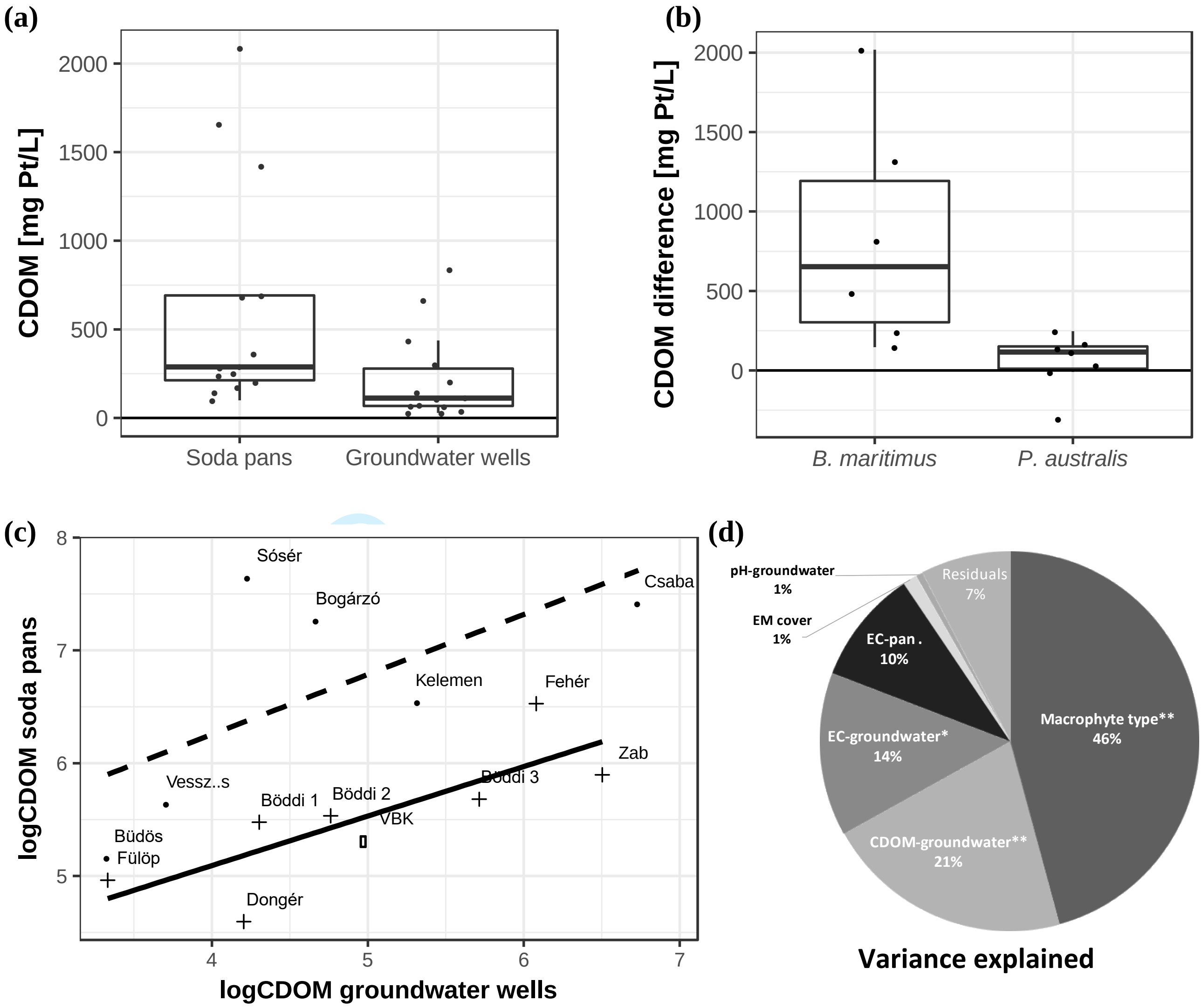


(b)



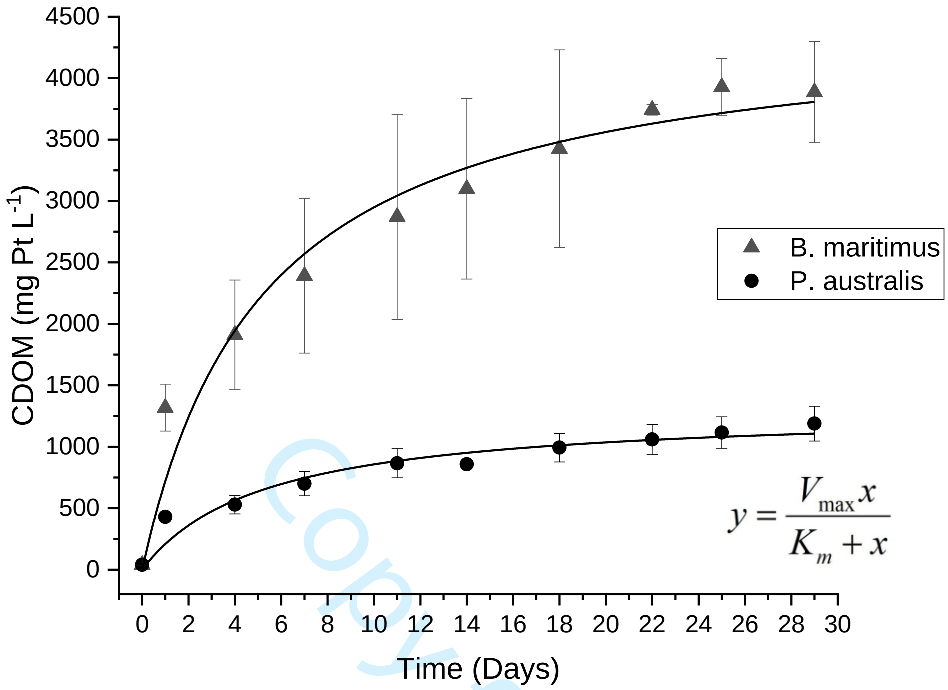

a)
DOC$_{mean}$ (mg L^{-1})
100
90
80
70
60
50
40
30
20
10
0
3,1
5
5,2
6,5
10
15
30
90
Deep lakes
Global lakes
N temp zone lakes
Surface water
Eutrophic lakes
Swamps, marshes
Bogs
Soda pans
b)
DOC$_{max}$ (mg L^{-1})
1000
900
800
700
600
500
400
300
200
100
0
60
160
300
988
Bogs
Coastal lagoons
Peatlands
Soda pans


N
AUSTRIA
River Danube
River Tisza
HUNGARY
Lake Balaton
Study area
Location in Europe
SERBIA
0
45
90 Km
Legend:
Soda pans
Main lakes and rivers
Alps and Carpathian Mountains

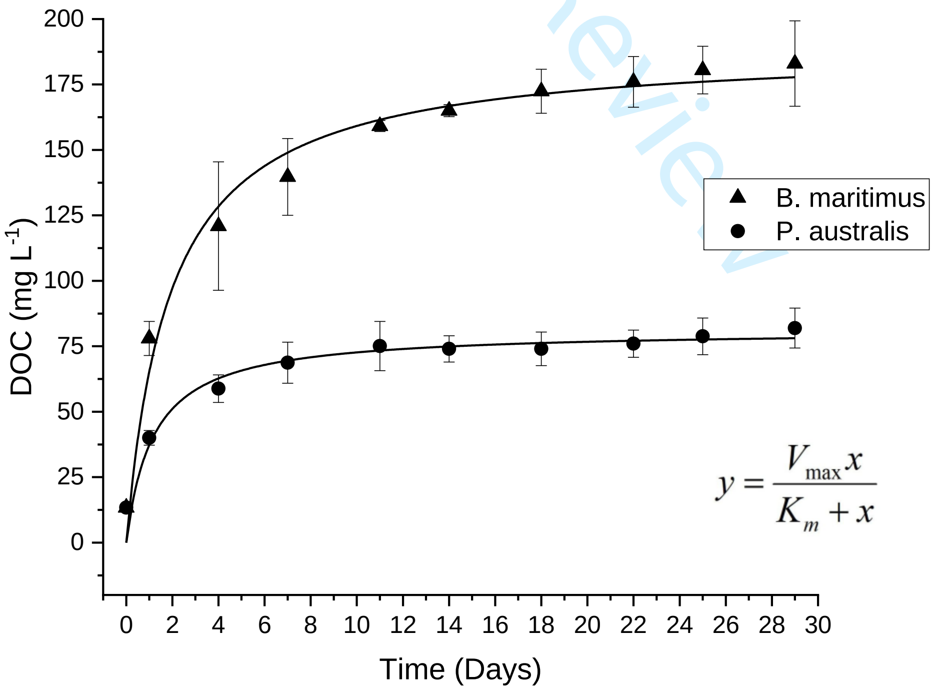


(a)
CDOM [mg Pt/L]
2000
1500
1000
500
0
Soda pans
Groundwater wells
(b)
CDOM difference [mg Pt/L]
2000
1500
1000
500
0
B. maritimus
P. australis
(c)
logCDOM soda pans
logCDOM groundwater wells
8
7
6
5
4
5
6
7
Sósér
Bogárzó
Csaba
Kelemen
Fehér
Zab
Vessz..s
Böddi 1
Böddi 2
Böddi 3
VBK
Büdös
Fülöp
Dongér
(d)
pH-groundwater 1%
EM cover 1%
Residuals 7%
EC-pan . 10%
EC-groundwater* 14%
Macrophyte type** 46%
CDOM-groundwater** 21%
Variance explained

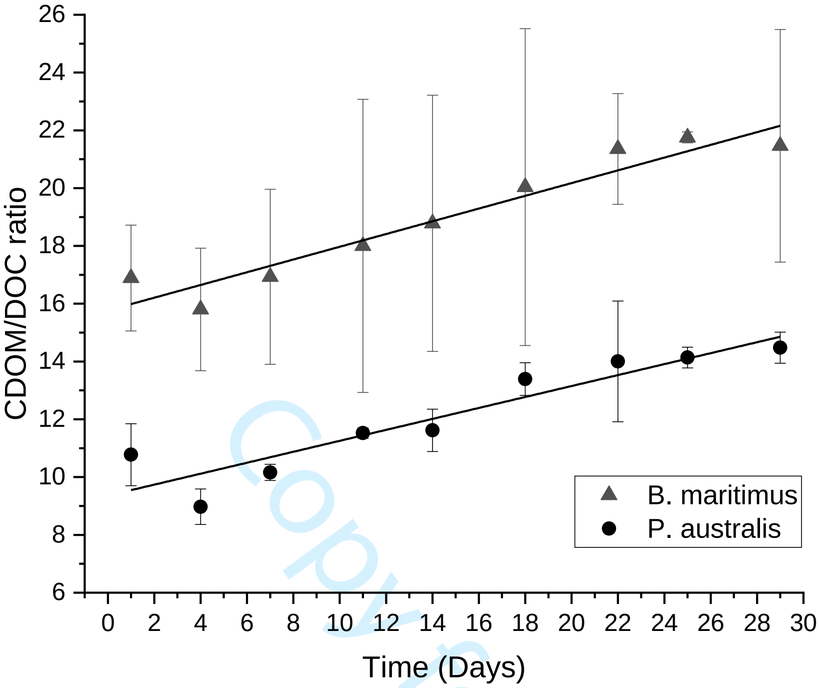
(a)

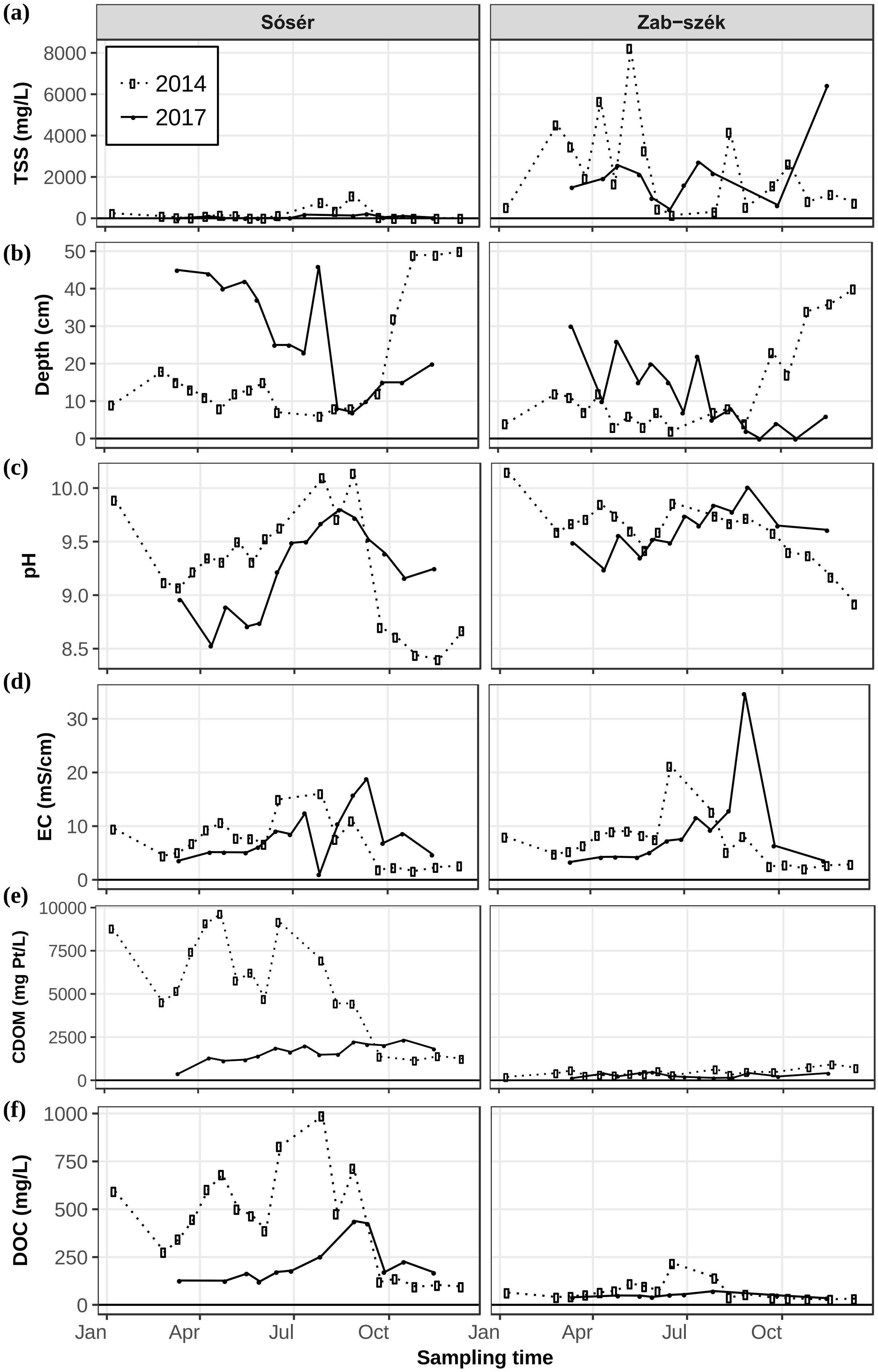

4500
4000
3500
3000
2500
2000
1500
1000
500
0
CDOM (mg Pt L-1)
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
Time (Days)
B. maritimus
P. australis
$y = \frac{V_{max} x}{K_m + x}$

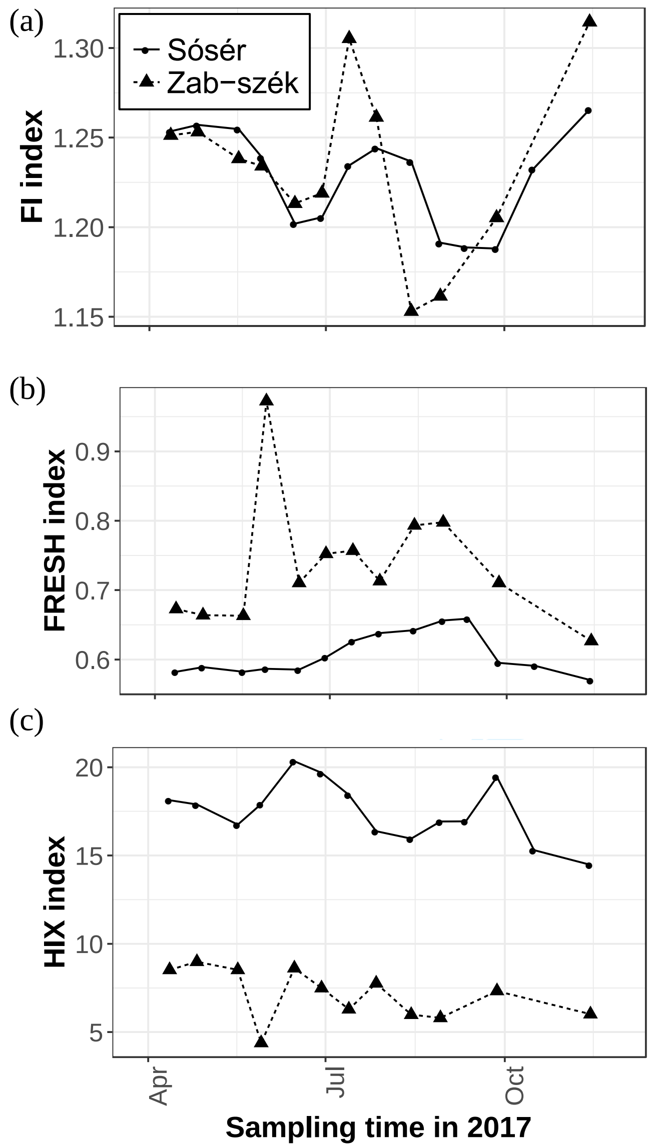
(b)


200
175
150
125
100
75
50
25
0
DOC (mg L-1)
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
Time (Days)
B. maritimus
P. australis
$y = \frac{V_{max} x}{K_m + x}$

(c)


CDOM/DOC ratio
26
24
22
20
18
16
14
12
10
8
6
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
Time (Days)
B. maritimus
P. australis


(a)
Sósér
Zab−szék
2014
2017
TSS (mg/L)
8000
6000
4000
2000
0
(b)
Depth (cm)
50
40
30
20
10
0
(c)
pH
10.0
9.5
9.0
8.5
(d)
EC (mS/cm)
30
20
10
0
(e)
CDOM (mg Pt/L)
10000
7500
5000
2500
0
(f)
DOC (mg/L)
1000
750
500
250
0
Jan
Apr
Jul
Oct
Sampling time


(a)
FI index
1.30
1.25
1.20
1.15
Sósér
Zab−szék
(b)
FRESH index
0.9
0.8
0.7
0.6
(c)
HIX index
20
15
10
5
Apr
Jul
Oct
Sampling time in 2017