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Spatiotemporal characterization of dissolved carbon for inland waters in semi-humid/semi-arid region, China

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Abstract. Spatiotemporal variations of dissolved organic carbon (DOC) and inorganic carbon (DIC) in 26 waters across the semi-humid/semi-arid Songnen Plain, China, were examined with data collected during 2008–2011. Fresh ($n=14$) and brackish ($n=12$) waters were grouped according to electrical conductivity (threshold = $1000\,\mu\mathrm{S\,cm^{-1}}$). Significant differences in the average DOC and DIC concentrations were observed between the fresh ($5.63\,\mathrm{mg\,L^{-1}}$, $37.39\,\mathrm{mg\,L^{-1}}$) and the brackish waters ($15.33\,\mathrm{mg\,L^{-1}}$, $142.93\,\mathrm{mg\,L^{-1}}$). Colored dissolved organic matter (CDOM) and DOC concentrations were mainly controlled by climatic–hydrologic conditions. The investigation indicated that the outflow conditions in the semi-arid region had condensed effects on the dissolved carbon, resulting in close relationships between salinity vs. DOC ($R^2=0.66$), and salinity vs. DIC ($R^2=0.94$). An independent data set collected in May 2012 also confirmed this finding (DOC: $R^2=0.79$, DIC: $R^2=0.91$), highlighting the potential of quantifying DOC and DIC via salinity measurements for waters dispersed in the plain. Indices based on the CDOM absorption spectra (e.g., the DOC-specific CDOM absorption (SUVA$_{254}$), absorption ratio $a_{250}:a_{365}(E_{250}:E_{365})$ and the spectral slope ratio (Sr, $S_{275-295}/S_{350-400}$)) were applied to characterize CDOM composition and quality. Our results indicate that high molecular weight CDOM fractions are more abundant in the fresh waters than the brackish waters.

1 Introduction

Dissolved organic matter (DOM) is one of the largest bioactive reservoirs on the earth's surface (Cole et al., 2007; Para et al., 2010). Though covering only a small fraction of the earth's surface, inland waters have a disproportional effect on the global carbon cycle (Cole et al., 1994, 2007; Tranvik et al., 2009; Armstrong, 2010), and play a vital role in burying, cycling and emitting carbon (Cole et al., 1994, 2007; Downing et al., 2008). DOM of inland waters, particularly its colored fraction (CDOM), influences light attenuation in waters (Vodacek et al., 1997; Zhang et al., 2007; Williamson and Rose, 2010; Stedmon et al., 2011), which in turn affects the transport and bio-availability of materials such as trace metals and organic pollutants (Cory et al., 2006; Schlesinger et al., 2011; Ledesma et al., 2012). The mineralization of dissolved organic carbon (DOC), the major component of DOM, is a source of CO_2 in the atmosphere (Cole et al., 2007; Duarte et al., 2008; Spencer et al., 2009; Tranvik et al., 2009). DOC also serves to mediate the chemical environment through organic acids (Wetzel, 2001; Tranvik et al., 2009) and to enhance or alleviate the toxic forms of heavy metals (e.g., aluminum or mercury) (Cory et al., 2006; Henneberry et al., 2011).

DOC mass balances are subject to the influences of temperature and precipitation, which can have important effects on DOC source, transport and fate (Larson et al., 2007; Jaffé et al., 2008; Fellman et al., 2011). Studies have indicated that concentrations of DOC in inland waters tend to decrease with increasing water residence times due to increased photobleaching and microbial activities (Curtis and Adams, 1995;

Helms et al., 2008; Julian et al., 2008, 2011; Stedmon et al., 2011). Several investigations have also illustrated that CDOM is inversely associated with salinity in estuarine and nearshore waters due to dominating riverine DOC inputs (Vodacek et al., 1997; Twardowski et al., 2004; Griffin et al., 2011). Inland waters in semi-arid to arid climates, however, generally tend to exhibit high salinity concentrations with elevated DOC concentrations (Brooks and Lemon, 2007). Curtis and Adams (1995) reported a positive correlation between DOC and specific conductivity in semi-arid east-central Alberta, Canada. However, this phenomenon, caused by evaporative concentration, has not been thoroughly investigated in other places (Wetzel, 2001; Brooks and Lemon, 2007). The role of saline water in DOC cycling needs further investigation (Duarte et al., 2008).

The source and composition of DOM can be investigated using various analytical approaches (Summers et al., 1987; Baker and Spencer, 2004; Spencer et al., 2008). These methods primarily dependent on the ultraviolet–visible (UV-Vis) and fluorescence measurements have been widely applied and reported (Chin et al., 1994; Vodacek et al., 1997; Helms et al., 2008; Zhang et al., 2007, 2010; Fellman et al., 2011; Stedmon et al., 2011). Using the ratio of the absorptions at 250 to 365 nm ($E_{250:365}$), De Han and De Boer (1987) successfully tracked changes in DOM molecule size, which was confirmed by other researchers (Peuravouri and Pihlaja, 1997; Zhang et al., 2007). The spectral slope (S) provides further information on the CDOM origin chemical processes and diagenesis, and the application of S is not limited by CDOM concentration (Twardowski et al., 2004; Helms et al., 2008; Fichot and Benner, 2011). Recently, Helms et al. (2008) developed another index based on the ratio of S value (Sr) at two narrow wavelength ranges: the S of the shorter wavelength region (275–295 nm) versus the longer wavelength region (350–400 nm). The effectiveness of Sr has been demonstrated with CDOM samples from various waters, ranging from DOC-rich wetlands to photobleached coastal waters and lakes over high-altitude plateaus (Helms et al., 2008; Zhang et al., 2010). Further, it has been modified to estimate DOC concentration using the CDOM absorption feature (Fichot and Benner, 2011; Spencer et al., 2012).

Dissolved inorganic carbon (DIC), largely composed of dissolved carbon dioxide and bicarbonate, is the primary source of carbon for photosynthesis and the generation of organic substances (Duarte et al., 2008). These inorganic compounds are generated by phytoplankton and higher plants in both lakes and rivers (Wetzel, 2001; Wilson and Xenopoulos, 2009), or they are generated externally in the drainage basin and imported to water bodies (Tranvik et al., 2009). The photobleaching process also produces DIC (Wetzel, 2001; Barros et al., 2011; Lapierre and Giogio, 2012). DIC is a major constituent of inland waters, which influences many characteristics of gaseous and nutrient availability and serves as an indicator of organic productivity. DIC also influences water quality properties such as acidity, hardness and related characteristics (Wetzel, 2001). Thus, it is necessary to evaluate the rudiments of DIC reactivity (Tranvik et al., 2009; C. C. Song et al., 2011).

As far as the role of DOM in carbon cycling is concerned, a number of key questions need to be addressed – for example, the origin of DOM, conversion of DOC to DIC, and CO_2 outgassing from various inland waters (Cole et al., 2007; Sobek et al., 2007; Jaffé et al., 2008; Tranvik et al., 2009). The spatiotemporal dynamics of dissolved carbon and its association with terrestrial inputs for inland aquatic ecosystems in semi-arid environments have only been investigated in a few regions (Brooks and Lemon, 2007; Mattsson et al., 2009; Moore et al., 2011). Further study is needed for characterizing dissolved carbon in inland saline waters (Duarte et al., 2008; Tranvik et al., 2009), and quantifying the role of inland saline waters for carbon cycling (Cole et al., 2007; Duarte et al., 2008). DOC and DIC concentrations in inland waters are regulated by a combination of parameters. While these parameters vary in both time and space within and across ecosystems, they can be captured by the analysis of optical properties. The objectives of this study were as follows: (1) to investigate the spatial variations of DOC and DIC in the semi-humid and semiarid region; (2) to examine the seasonal characteristics of DOC and DIC; and (3) to perform a DOM source analysis using the relationship between optical absorption indices and salinity.

2 Materials and methods

2.1 Study area

The Songnen Plain is located in the central region of northeast China, covering an area of approximate 22.3×10^4 km^2 (42°49′–49°12′ N, 121°38′–128°30′ E; Fig. 1). It is a fluvial plain formed by the Songhua and Nenjiang rivers and their tributaries originating from the surrounding mountains: the Changbai (east), Lesser Xing’an (north) and Great Xing’an (west) mountain ranges. The plain is characterized by a temperate, semi-humid and semi-arid continental monsoon climate, with seasons alternating between dry and windy spring; humid and warm summer; windy and dry autumn; and long, cold and dry winter (Song et al., 2010; Zeng et al., 2011). The air temperature increases from north to south, spanning from 2 to 6 °C. The average annual precipitation ranges from 350 to 600 mm, of which more than 80 % of the precipitation occurs in growing season (from May to October). The potential evaporation exceeds 1300 mm yr^{-1}, resulting in water scarcity. Due to its geomorphology, many terminal-flow areas and temporary waters are formed, which result in widely distributed fresh and saline water bodies in the plain. These lakes in mid-west of the plain were formed in a similar geological and climatic environment (K. S. Song et al., 2011), and waters tend to be brackish. Particularly, these terminal waters have no outflow (Table 1) due to saline and

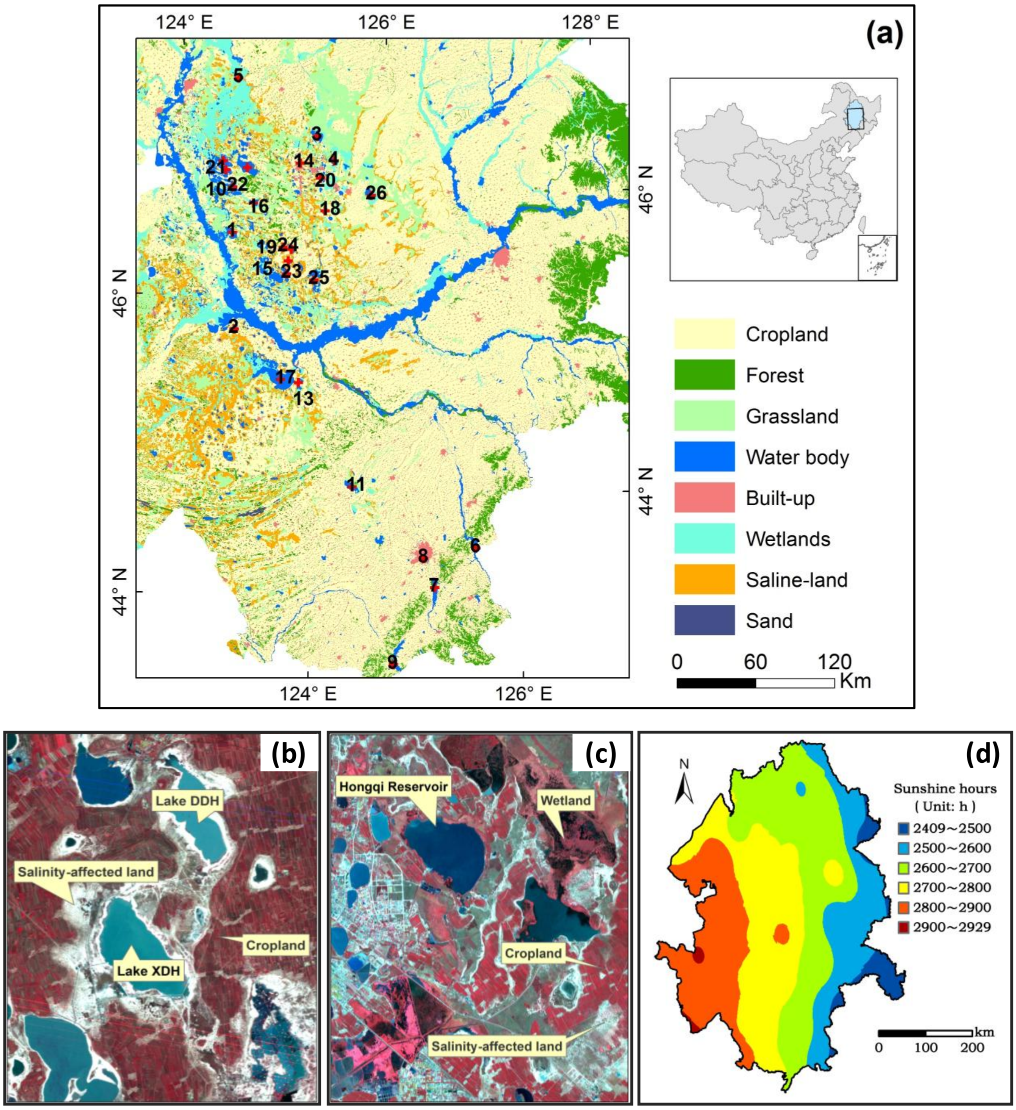

Fig. 1. (a) Map of sampling locations with various land use/land cover types. The fresh waters are the following: 1 – Lamasi, 2 – Yueliang Lake, 3 – Daqing Reservoir, 4 – Hongqi Reservoir, 5 – Dongsheng Reservoir, 6 – Shitoukoumen Reservoir, 7 – Xinlicheng, 8 – Nanhu Lake, 9 – Erlonghu, 10 – Talahong Reservoir, 11 – Boluo Lake, 12 – Longhupao Lake, 13 – Xinmiao Lake, 14 – Yueliangpao; brackish waters are the following: 15 – Xidahai, 16 – Yuebingpao, 17 – Chagan Lake, 18 – Zhongneipao, 19 – Dongdahai, 20 – Zhaojiatun, 21 – Cuibaguzi, 22 – Huoshaohei, 23 – Nanyin Reservoir, 24 – Xinhua Lake, 25 – Kulipao, and 26 – Qingkenpao. **(b)** Lake Dongdahai and Xidahai, **(c)** Hongqi Reservoir with their ambient environments, and **(d)** sunshine duration characteristics for the Songnen Plain.

alkaline soil erosion (Fig. 1a–c). A total of 14 fresh (open water) waters and 12 brackish (terminal) waters were chosen for this study (threshold value = 1000 μS cm^{-1} was set to group fresh and brackish waters, Table 1) with respect to both climatic–hydrologic conditions and lake size. Note that Nanhu Lake (NHL) is located in Changchun City, and some sewage outlets are connected with it.

2.2 Water sampling and quality determination

A total of 26 water bodies were sampled from late August to early October 2011 for spatially characterizing dissolved carbon. Altogether, 211 samples were collected, and the sample numbers for each water body are listed in Table 1. Six field surveys were conducted across the Shitoukoumen Reservoir (fresh water: 109 samples) and Chagan Lake (brackish water: 107 samples) respectively to examine the seasonal characteristics during 2008–2011. Surface water samples were collected at each station approximately 0.5 m below the water surface, generally located in the middle of water bodies. All water samples were kept in a portable refrigerator powered by field trip vehicles. All samples were filtered within 24 h after coming back from the field.

Table 1. Lake (or reservoir) names, abbreviation (with numbers), sampling dates, mean water surface area, water volume, water depth, inflow and outflow conditions, pH value, salinity and sampling numbers for waters across the Songnen Plain.

Lake name	Abbr. (number)	Date	Area (km^2)	Volume (10^8 m)	Depth (m)	In-F[a]	Out-F[b]	pH	Salinity (PSU)	*N*
Lamasi	LMS (1)	24 September 2011	51	1.52	3	P	Y	8.46	0.09	5
Yueliang Lake	YLL (2)	1 September 2011	206.1	4.74	3.6	R + P	Y	7.93	0.1	20
Daqing	DQR (3)	8 September 2011	56.1	1.03	1.3	C + P	Y	8.14	0.1	5
Hongqi	HQR (4)	9 September 2011	26.2	0.83	2.8	C + P	Y	8.36	0.11	3
Dongsheng	DSR (5)	25 September 2011	7.8	0.31	3.2	C + P	Y	8.05	0.11	5
Shitoukoumen	STR (6)	2008–2010 #	73.3	3.86	7.5	R + P	Y	8.09	0.13	15
Xinlicheng	XLC (7)	20 September 2011	58.5	5.92	7.6	R + P	Y	8.57	0.14	20
Nanhu	NHL (8)	26 September 2011	1	0.03	3.7	SW + R	Y	8.28	0.15	8
Erlonghu	ELL (9)	19 September 2011	89.5	1.76	11.6	R + P	Y	8.23	0.16	21
Talahong	TLR (10)	16 September 2011	71.6	1.89	1.8	R + P	Y	8.64	0.24	4
Boluohu	BLL (11)	25 August 2011	49.3	0.9	1.3	R + P	Y	8.6	0.25	8
Longhupao	LHP (12)	23 September 2011	130.8	1.75	2.7	R + P	Y	8.09	0.37	10
Xinmiao	XML 13)	30 August 2011	31.6	0.62	2.1	C + P	Y	9.12	0.46	10
Yueliangpao	YLP (14)	7 September 2011	1.1	0.07	1.5	R + P	N	7.78	0.48	3
Xidahai	XDH (15)	14 September 2011	12.5	0.46	1.6	P	N	8.97	0.52+	3
Yuebingpao	YBP (16)	22 September 2011	23	0.57	2.5	P	N	8.92	0.54+	3
Chagan	CGL (17)	2008–2011#	375.2	6.42	2.2	C + P	N	9.12	0.57+	15
Zhongneipao	ZNP (18)	10 September 2011	13.6	0.55	1.7	C + P	N	9.09	0.65+	10
Dongdahai	DDH (19)	14 September 2011	16.5	0.25	0.4	P	N	8.91	0.66+	5
Zhaojiatun	ZJT (20)	8 September 2011	5.6	0.17	0.7	P	N	8.42	0.68+	4
Cuibaguzi	CBG (21)	25 September 2011	58.4	1.12	1.4	P	N	9.35	0.71+	1
Huoshaohei	HSH (22)	17 September 2011	73.4	1.92	2.5	P	Y	9.28	0.73+	4
Nanyin	NYR (23)	15 September 2011	47.1	1.05	2.1	C + P	N	9.17	0.95+	4
Xinhuahu	XHL (24)	14 September 2011	7.4	0.18	1.3	P	N	9.33	1.25+	3
Kulipao	KLP (25)	15 September 2011	33.7	0.71	2.1	R + P	N	9.27	1.42+	5
Qingkenpao	QKP (26)	11 September 2011	33.1	0.73	1	C + P	N	9.33	1.51+	5

[a] denotes the major inflow type, R = river, C = channel, P = precipitation, SW = sewage; [b] denotes the out flow type, Y = yes, N = no; # indicates that multiple field campaigns were conducted across these waters; + indicates brackish waters.

Water turbidity was determined using a Shangfen Vis-7230 spectrophotometer with a 3 cm quartz cell at room temperature (20 ± 2°) with Milli-Q water as reference. Salinity was measured through DDS-307 electrical conductivity (EC) meter with $\mu\mathrm{S\,cm^{-1}}$ (micro-Siemens/centimeter) units at room temperature (20±2°) in the laboratory. Chlorophyll *a* (Chl *a*) concentration was determined using a Shimadzu UV-2550PC spectrophotometer (K. S. Song et al., 2011). Total suspended matter (TSM) was determined gravimetrically; details can be found in Song et al. (2013). Total nitrogen (TN) was measured based on the absorption levels at 146 nm of water samples decomposed with alkaline potassium peroxydisulfate. Total phosphorus (TP) was determined using the molybdenum blue method after the samples were digested with potassium peroxydisulfate (APHA, 1998). To determine DOC concentrations, water samples were filtered through pre-combusted 0.45 µm GF/F filters. The standards for dissolved total carbon (DTC) were prepared from reagent grade potassium hydrogen phthalate in ultra-pure water, while DIC levels were determined using a mixture of anhydrous sodium carbonate and sodium hydrogen carbonate. DOC was calculated by subtracting DIC from DTC, both of which were measured by high-temperature catalytic oxidation (680 °C) using a total organic carbon analyzer (Shimadzu, TOC-VCPN). To validate the association between salinity and DOC and DIC, 153 independent samples across 26 water bodies were analyzed in May, 2012.

2.3 CDOM laboratory analysis

In laboratory, samples were filtered at low pressure through pre-combusted GF/F filters, and then through a pre-rinsed Millipore membrane cellulose filter (0.22 µm) for CDOM absorption measurement. Absorption spectra were obtained in the 200 to 800 nm spectral region at 1 nm increments using a Shimadzu UV-2550PC UV-Vis dual beam spectrophotometer with 1cm quartz cuvettes. Milli-Q water was used as reference for CDOM absorption measurements. The absorption coefficient (a_{CDOM}) was calculated from the measured optical density (OD) of the sample using Eq. (1):

$$a_{\mathrm{CDOM}}(\lambda) = 2.303[\mathrm{OD}_{S(\lambda)} - OD_{(\mathrm{null})}]/\gamma, \qquad (1)$$

where γ is the cuvette path length (0.01 m) and 2.303 is the conversion factor. Some fine particles may have remained in the filtrate and necessitated correction for scattering by fine particles (Babin et al., 2003). $\mathrm{OD}_{(\mathrm{null})}$ is the average optical density over 740–750 nm, where the absorbance of CDOM was assumed to be zero. All absorption measurements were conducted within 48 h of field sampling. The CDOM absorption ratio, $E_{250:365}$, was calculated using absorbance at 250 nm and 365 nm. The specific UV absorbance at 254 nm (SUVA_{254}) is defined as the absorbance at 254 nm (m^{-1}) divided by the DOC concentration ($\mathrm{mg\,L}^{-1}$) (Weishaar et al., 2003).

2.4 Spectral slope (*S*)

A CDOM absorption spectrum, $a_{\mathrm{CDOM}}(\lambda)$, can be expressed as an exponential function (Bricaud et al., 1995; Babin et al., 2003):

$$a_{\mathrm{CDOM}}(\lambda_i) = a_{\mathrm{CDOM}}(\lambda_\mathrm{r})\exp[-S(\lambda_i - \lambda_\mathrm{r})], \tag{2}$$

where $a_{\mathrm{CDOM}}(\lambda_i)$ is the CDOM absorption at a given wavelength λ_i, $a_{\mathrm{CDOM}}(\lambda_\mathrm{r})$ is the absorption estimate at a reference wavelength λ_r (440 nm), and S is the spectral slope. S is calculated by fitting the data to a nonlinear model over a wavelength range of 300 to 500 nm, as suggested by Zepp and Schlotzhauer (1981) and Zhang et al. (2007). Different S values may be obtained due to different curve-fitting approaches and spectral ranges (Babin et al., 2003; Binding et al., 2008; Astoreca et al., 2009). The Sr was calculated to determine CDOM sources (Helms et al., 2008).

2.5 Statistical analysis

Statistical analyses were conducted using SPSS 16.0 software package (Statistical Program for Social Sciences). The differences in DOC, DIC, CDOM and relevant spectral indices were assessed with ANOVA (paired t test). Both p value (α) and statistical power (power = 1-β) were calculated for a comparison of brackish and fresh waters in the plain.

3 Results

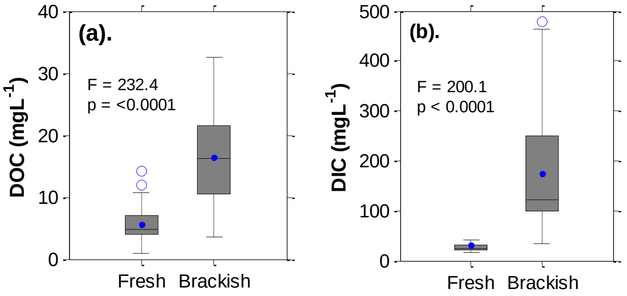

Fig. 2. Box plots of DOC **(a)** and DIC **(b)** for fresh and brackish waters in the Songnen Plain. The black line and the blue filled circles represent the median and mean respectively. The horizontal edges of the boxes denote the 25th and 75th percentiles; the whiskers denote the 10th and 90th percentiles and the blue circles represent outliers.

The analysis of the water samples collected from the 26 water bodies in the Songnen Plain indicated that these water bodies had quite diverse water qualities. Most of these water bodies exhibited high TN concentrations (mean ± SD, $3.25 \pm 1.86\,\mathrm{mg\,L}^{-1}$), and only two lakes contained average TN levels less than $1.0\,\mathrm{mg\,L}^{-1}$ (Table 2). Similarly, TP levels exhibited high variability, ranging from 0.06 to $1.86\,\mathrm{mg\,L}^{-1}$ ($0.44 \pm 0.52\,\mathrm{mg\,L}^{-1}$). As expected, high Chl a concentrations ($48.44 \pm 39.71\,\mathrm{\mu g\,L}^{-1}$) were observed in these water bodies. According to Carlson's trophic index (1977), 85 % of the water bodies were eutrophic or hypereutrophic (Zhang et al., 2010), and the remaining 15 % were mesotrophic. Agricultural non-point pollution was the main source of TN and TP for these water bodies, resulting in algal blooms in some of the waters (Wang et al., 2009; K. S. Song et al., 2011). Due to regional hydro-geologic and climatic conditions, most of water bodies in the western Songnen Plain had high salt contents and pH values. Overall, these water bodies were highly turbid (87.0 ± 101.4 NTU), with a high concentration of TSM ($119.55 \pm 131.37\,\mathrm{mg\,L}^{-1}$) in the water column (Table 2). This could be attributed to strong winds in spring and autumn causing re-suspension of the bottom sediments.

3.1 Dissolved carbon spatial characteristics

The DOC concentrations in fresh waters ranged from $1.01\,\mathrm{mg\,L}^{-1}$ in Longhupao Lake (LHP) to $14.23\,\mathrm{mg\,L}^{-1}$ in Boluo Lake (BLL) (see Table 2). A large variation of DOC concentration was revealed in the brackish waters, ranging from $3.6\,\mathrm{mg\,L}^{-1}$ in Nanyin Reservoir (NYR) to $32.6\,\mathrm{mg\,L}^{-1}$ in Zhongneipao (ZNP). The brackish waters exhibited higher average DOC concentrations ($16.4 \pm 7.4\,\mathrm{mg\,L}^{-1}$) than fresh waters ($5.6 \pm 2.4\,\mathrm{mg\,L}^{-1}$). A significant difference was observed between these two water types ($F = 232.4$, $p < 0.0001$, power = 1.0, Fig. 2a), and the interquartile ranges (IQR) for the fresh and brackish waters are 2.91 and 11.31, respectively. A large variation of DIC concentrations in the fresh waters was observed, ranging from $16.2\,\mathrm{mg\,L}^{-1}$ over Shitoukoumen Reservoir (STR) in eastern part of the plain to $125.4\,\mathrm{mg\,L}^{-1}$ in Huoshaohei (HSH) in western Songnen Plain (Fig. 1). The DIC concentrations for the brackish waters ranged from $37.2\,\mathrm{mg\,L}^{-1}$ (ZJT) to $482.4\,\mathrm{mg\,L}^{-1}$ (DDH). Concentrations of DIC were higher in the brackish waters ($173.5 \pm 110.4\,\mathrm{mg\,L}^{-1}$) than the fresh water ($32.0 \pm 24.3\,\mathrm{mg\,L}^{-1}$). A significant difference for DIC was observed between the brackish and fresh waters ($p < 0.0001$, power = 1.0, Fig. 2b). Higher IQR values were recorded with brackish water (151.07) than fresh water (9.53). Interestingly, the fresh waters showed a higher

Table 2. Mean values of water quality parameters and CDOM absorption from late August to late September. TN and TP denote total nitrogen and total phosphorus, respectively. Chl *a* denotes chlorophyll *a* concentration. TSM denotes total suspended matter, and Turb denotes water turbidity. DOC and DIC are dissolved organic and inorganic carbon respectively. "–" indicates no available data.

Lake Abbr	TN (mg L^{-1})	TP (mg L^{-1})	Chl *a* (µg L^{-1})	TSM (mg L^{-1})	Turb (NTU)	a_{CDOM} (355) (m^{-1})	a_{CDOM} (440) (m^{-1})	DOC	DIC
LMS	0.96	0.02	17.6	39.9	52.1	5.04	1.46	9.15	23.42
YLL	9.77	1.86	55.7	44	51.4	25.45	10.39	8.79	25.48
DQR	2.08	0.07	11.7	40.2	37.6	5.69	1.48	5.23	30.13
HQR	5.28	1.01	18.1	17.7	24.3	7.56	1.57	5.71	28.05
DSR	5.4	1.72	136.4	44.6	22.4	16.91	4.25	5.55	30.84
STR	1.6	0.06	23.4	35.7	37.8	2.88	0.77	4.74	16.23
XLC	0.89	0.06	14.3	8.1	10.7	3.08	0.82	3.83	21.13
NHL	2.15	0.14	20.2	80.4	10.3	8.66	2.53	5.08	20.15
ELL	1.94	0.13	60.7	14	7.3	3.45	0.96	4.21	22.59
TLR	4.42	1.21	52	554.6	97.2	11.9	3.06	6.36	74.59
BLL	4.37	1.13	48.5	402.6	44.8	31.33	15.22	9.69	88.98
LHP	3.5	0.48	59.9	258.9	64.5	26.32	10.65	2.72	33.43
XML	–	–	28.5	80.8	21.2	9.04	2.84	4.15	23.17
YLP	2.51	0.57	9.4	29.5	23.9	11.17	2.3	6.36	74.58
XDH	1.91	0.03	31.7	20.9	366.8	7.35	1.38	14.92	244.53
YBP	2.41	0.2	43.1	63.3	149.6	3.64	0.52	19.97	111.53
CGL	2.78	0.43	46.3	65.7	88.5	8.37	1.69	9.98	98.25
ZNP	2.17	0.1	14.1	110.3	27.8	6.91	1.33	29.78	124.36
DDH	4.9	0.13	185.7	45.2	408.5	8.77	1.97	28.44	442.78
ZJT	4.45	0.17	96.8	143.3	11.7	16.64	5.56	14.44	82.11
CBG	1.09	0.079	35.2	23.8	264.3	5.96	2.28	18.57	233.75
HSH	1.82	0.075	39.7	25.2	132.4	9.49	4.13	7.33	125.35
NYR	1.04	0.55	29.4	5.6	124.3	2.05	0.75	6.85	54.81
XHL	3.75	0.24	33.5	113.8	47.2	3.06	0.43	14.92	244.50
KLP	5.96	0.41	88	311	23.4	15.37	5.23	16.86	106.45
QKP	4.18	0.15	59.5	129.3	112.2	6.92	1.29	20.56	285.23

mean DOC/DIC ratio (0.33 ± 0.13) than the brackish water (0.21 ± 0.14).

3.2 CDOM spatial characterization

As shown in Table 2, the mean value of SUVA_{254} ranged from 2.3 L mg C^{-1} m^{-1} (±0.14 SD) in Nanhu Lake (NHL) to 8.7 L mg C^{-1} m^{-1} (±2.8 SD) in Huoshaohei (HSH). The small mean SUVA_{254} value was observed in the brackish waters, ranging from 2.8 L mg C^{-1} m^{-1} (±0.06 SD) in the Chagan Lake (CGL) to 5.7 L mg C^{-1} m^{-1} (±0.98 SD) in the Yuebingpao (YBP). This indicates a significant difference in SUVA_{254} between the fresh and brackish waters (Fig. 3a, $p < 0.0001$, power = 1.0). The IQR values are 2.00 and 1.41 for the fresh and brackish waters, respectively. The averaged SUVA_{254} was higher in the fresh waters (5.8 L mg C^{-1} m^{-1} ± 1.72 SD) than the brackish waters (3.8 ± 1.04 L mg C^{-1} m^{-1}).

The averaged $E_{250:365}$ ranged from 5.5 (±1.46 SD) in Xinmiao Lake (XML) to 10.3 (±0.27 SD) in Huoshaohei (HSH) for the fresh water (Table 2), but a large range for $E_{250:365}$ was obtained for the brackish waters, ranging from 7.5 ± 0.10 in Kulipao (KLP) to 15.8 ± 1.43 in Xinmiao Lake (XML). This suggests a significant difference ($p < 0.0001$, power = 0.999, Fig. 3b) between the fresh and the brackish waters in $E_{250:365}$. IQR for the brackish water (5.67) was higher than the fresh water (1.17).

As shown in Table 2, the averaged Sr for the fresh waters ranged from 0.80 ± 0.021 in Daqing Reservoir (DQR) to 1.46 ± 0.046 SD in Nanhu Lake (NHL). Comparatively, a larger range for Sr was obtained for the brackish waters, ranging from 1.32 (±0.006 SD) in Kulipao (KLP) to 2.1 (±0.123 SD) in Xidahai Lake (XDH). The Sr values exhibited an obvious difference for the fresh and brackish waters (Fig. 3c, $p < 0.0001$, power = 0.999), in which the averaged Sr values were 1.09 ± 0.17 and 1.41 ± 0.32 for the fresh and brackish waters, respectively. IQR value for the fresh water was lower (0.21) than that for the brackish water (0.36).

3.3 Seasonal variation

3.3.1 Dissolved carbon

The water quality characteristics in both STR and CGL are shown in Table 3. Apparent seasonal variations were observed for TN, TP, Chl *a* and TSM concentration in both

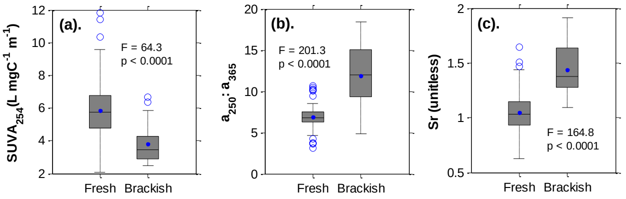

Fig. 3. Box plots of **(a)** SUVA_{254}, **(b)** CDOM ratio ($a_{250} : a_{365}$) and **(c)** spectral slope ratio (Sr: $S_{275-295} : S_{350-400}$ nm) between fresh and brackish waters in the Songnen Plain. The black line and the blue filled circles represent the median and mean respectively. The horizontal edges of the boxes denote the 25th and 75th percentiles; the whiskers denote the 10th and 90th percentiles and the blue circles represent outliers.

water bodies. Salinity, pH, and the CDOM absorption for the CGL were observed higher than that those for the STR (Table 3). As shown in Fig. 4a, the DOC concentrations in the STR were lower than the CGL in the growing season. Higher DOC concentrations were observed in May for both the STR and CGL, and lower DOC concentrations were observed from May to October in the STR (Fig. 4a). In contrast, the CGL has higher DOC concentrations in the May and October, and lower values in July, August and September for (Fig. 4a).

As illustrated in Fig. 4b, the overall DIC concentration in the CGL was significantly higher than that from the STR ($F = 337.2$, $p < 0.0001$). However, higher DIC was observed in May for the STR, which is consistent with the DOC concentration illustrated in Fig. 4a. Likewise, a higher DIC variation was observed in May for the STR, but small variation values were observed for the other months. The CGL showed high DIC concentrations with less seasonality, except for June and August (Fig. 4b). The samples in June and August revealed a relatively large variation, but small variations in September and October.

3.3.2 CDOM temporal characterization

As illustrated in Table 3, the CGL exhibited significantly higher a_{CDOM} (355) values ($17.5 \pm 12.03\,\mathrm{m}^{-1}$) than the STR ($5.6 \pm 0.72\,\mathrm{m}^{-1}$; $p < 0.003$). Likewise, significant differences in the a_{CDOM} (440) were found between the CGL and STR ($p < 0.001$). As shown in Fig. 5a, the STR and CGL exhibited higher SUVA_{254} values in May, but in other months the SUVA_{254} in the CGL showed smaller values and less variation than that in the STR (Fig. 5a). Overall, a significant difference of SUVA_{254} between the fresh and brackish waters during ice-free season was observed ($p < 0.0001$, power = 0.999)

While $E_{250:365}$ generally increased from May to October in the CGL (Fig. 5b), $E_{250:365}$ values for the STR were relatively stable through all months except for May. Low $E_{250:365}$ values were recorded in May and June for the CGL.

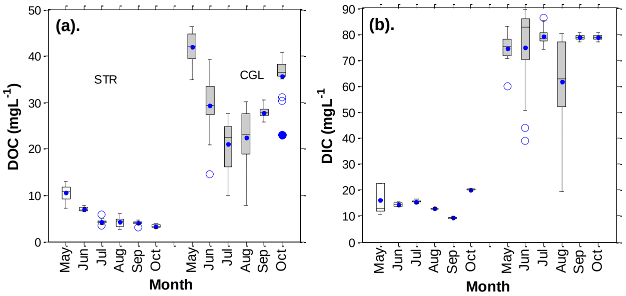

Fig. 4. Box plots of dissolved carbon in fresh and brackish waters during ice-free season: **(a)** DOC and **(b)** DIC for Shitoukoumen Reservoir (STR) and Chagan Lake (CGL). The black line and the blue filled circles represent the median and mean respectively. The horizontal edges of the boxes denote the 25th and 75th percentiles; the whiskers denote the 10th and 90th percentiles and the blue circles represent outliers.

Overall increased Sr values were observed in the STR and CGL (Fig. 5c). It can be seen that the Sr values followed similar trends as shown in $E_{250:365}$ for both the STR and CGL, while inverse trends were observed for the SUVA_{254} values. Although a significant difference of $E_{250:365}$ was revealed between the fresh and brackish waters (Fig. 5b), the difference was not as significant as those demonstrated for SUVA_{254} (Fig. 5a) and Sr (Fig. 5c).

3.4 Dissolved carbon versus salinity

As stated above, higher DOC and DIC concentrations were illustrated in the brackish waters in the Songnen Plain with high salinity concentrations due to the terminal aquatic environments, representing terminal points for carbon flow in the terrestrial landscape (Duarte et al., 2008). Regression analyses between salinity and dissolved carbon parameters were conducted on the samples collected in 2011 from 26 water bodies (Fig. 6). The concentrations of DOC in various waters increased with salinity (Fig. 6a; $R^2 = 0.66$, $p < 0.001$). Likewise, a linear relationship between DIC and salinity was observed (Fig. 6b; $R^2 = 0.94$, $p < 0.0001$). To test the stability of the relationship, the correlation of DOC to salinity for the samples in May 2012 is demonstrated in Fig. 6c ($R^2 = 0.96$, $p < 0.0001$). Even when four samples with extremely high values (from XDH, see Fig. 1b) were eliminated, a high determination coefficient was still achieved ($y = 67.25x + 11.216$; $R^2 = 0.79$, $p < 0.0001$). Similarly, a high correlation between DIC and salinity existed (Fig. 6d; $R^2 = 0.98$, $p < 0.0001$). A high R^2 value was also obtained between DIC and salinity ($y = 194.74x + 9.66$; $R^2 = 0.91$, $p < 0.0001$) when four samples with extremely high values were excluded.

Table 3. Seasonal variability of water quality parameters and CDOM absorption features for the Shitoukoumen Reservoir (STR) and for Chagan Lake (CGL); all water quality parameters have the same units as shown in Table 2.

	Date	TN	TP	Chl *a*	TSM	Salinity	pH	a_{CDOM} (355)	a_{CDOM} (440)	*N*
STR 1	5/20/09	2.80	0.14	12.06	60.15	–	7.86	4.34	1.71	17
STR 1	6/13/08	1.25	0.06	24.25	21.77	0.13	8.53	6.15	2.84	18
STR 3	7/27/09	1.03	0.12	6.98	14.51	0.13	8.43	5.32	2.06	22
STR 4	8/29/09	0.99	0.07	7.32	38.51	0.12	8.31	5.82	2.46	25
STR 5	9/23/08	1.09	0.08	35.23	23.79	0.13	8.54	5.96	2.28	20
STR 6	10/07/10	1.17	0.09	18.7	18.7	0.13	8.71	4.57	1.83	7
Subtotal	–	1.35	0.09	17.4	35.80	0.13	8.42	5.56	2.28	109
CGL 1	5/03/11	2.06	0.073	6.34	57.55	0.29	8.63	26.13	8.84	18
CGL 2	6/14/08	2.01	0.13	11.6	43.2	0.37	9.07	13.5	6.87	8
CGL 2	7/15/09	3.28	0.24	14.31	190.12	0.76	9.61	34.02	19.74	20
CGL 3	8/29/09	2.21	0.182	9.82	233.08	0.48	9.10	10.26	4.64	19
CGL 4	9/13/10	1.82	0.075	39.67	25.19	0.66	9.30	9.49	4.13	25
CGL 5	10/12/09	1.82	0.054	4.58	66.81	0.67	9.27	6.76	2.73	17
Subtotal	–	2.24	0.125	14.95	114.52	0.57	9.20	17.50	7.93	107

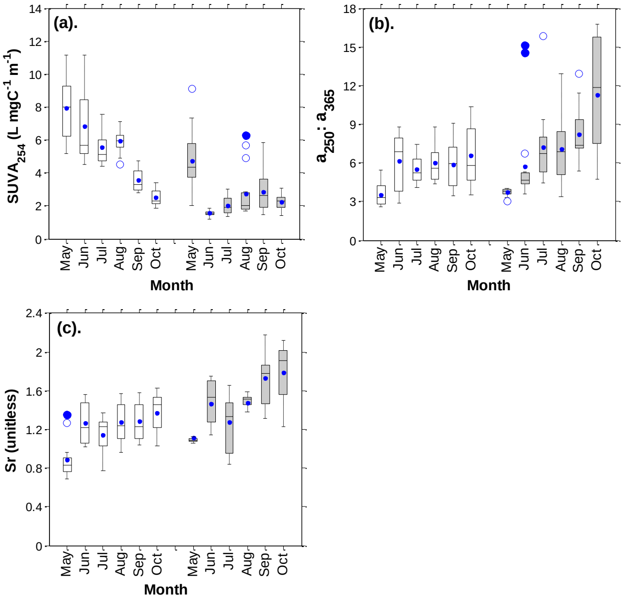

Fig. 5. Box plots of **(a)** SUVA_{254}, **(b)** $a_{250}:a_{365}$ and **(c)** Sr for Shitoukoumen Reservoir (empty box) and Chagan Lake (filled box) in various seasons. The black line and the blue filled circles represent the median and mean respectively. The horizontal edges of the boxes denote the 25th and 75th percentiles; the whiskers denote the 10th and 90th percentiles and the blue circles represent outliers.

4 Discussion

4.1 Dissolved organic carbon in fresh and brackish waters

In this study, substantial variations for both DOC and DIC are observed between the fresh and brackish waters in the Songnen Plain (Fig. 2). The pattern is similar to that reported by Curtis and Adams (1995) for lakes in east-central Alberta in the semi-arid region of Canada. Seasonally averaged DOC concentrations in the brackish waters are much higher than in the fresh water bodies, indicating that DOC concentration violates the trend in humid regions, with DOC decreasing with water residence time due to prolonged photobleaching and possible dilution (Twardowski and Donaghay, 2002; Julian et al., 2011; Spencer et al., 2012). These findings indicate that regional hydrogeologic and climatic conditions play an important role in driving DOC variability (Sobek et al., 2007; Duarte et al., 2008; Jaffé et al., 2008). In the brackish waters, DOC accumulates via runoff passing through various landscapes (Larson et al., 2007; C. C. Song et al., 2011), and can be lost from these brackish waters through sinking to the bottom (Cole et al., 2007; Tranvik et al., 2009; Barros et al., 2011) or being transformed into DIC (includes $\mathrm{CO_2}$) in the water column (Brooks and Lemon, 2007; Duarte et al., 2008; Tranvik et al., 2009). Consequently, DOC and DIC accumulate in brackish (terminal) waters at much higher rates than those in fresh (open) waters (Brooks and Lemon, 2007; Duarte et al., 2008).

The DIC concentrations in the brackish waters in the western Songnen Plain are much higher than in the fresh waters in the eastern part of the plain. Comparatively shorter sunlight durations (Zeng et al., 2011) and water residence times (Wang and Dou, 1998) result in lower DIC production and accumulation in fresh waters. As noted, most of the waters in the western Songnen Plain exhibit the limnetic characteristics of shallow plain lakes, which is re-suspension of the loose bottom sediment due to strong wind (see Table 2 for the Kulipao Lake, Nanyin Reservoir and Cuibaguzi Lake). A long sunlight duration in the western part of the Songnen Plain may cause higher DIC concentrations due to enhanced photochemical oxidation processes (Brook and Lemon, 2007; Tobias and Bohlke, 2011), speeding up the

Table 4. Multi-regression analysis of DOC and DIC against accumulated climatic variables in one month before field surveys were conducted (P, precipitation; ET_0, potential evaporation; S, sunshine duration hours) over 26 water bodies across the Songnen Plain.

		Slope			Statistics			
	Intercept	P	ET_0	S	*R* square	Adj.R square	*F* value	*p* value
DOC	−135.38	0.18	0.93	0.11	0.33	0.27	4.13	0.024
DIC	−1823.77	2.89	0.71	5.99	0.34	0.26	3.78	0.032

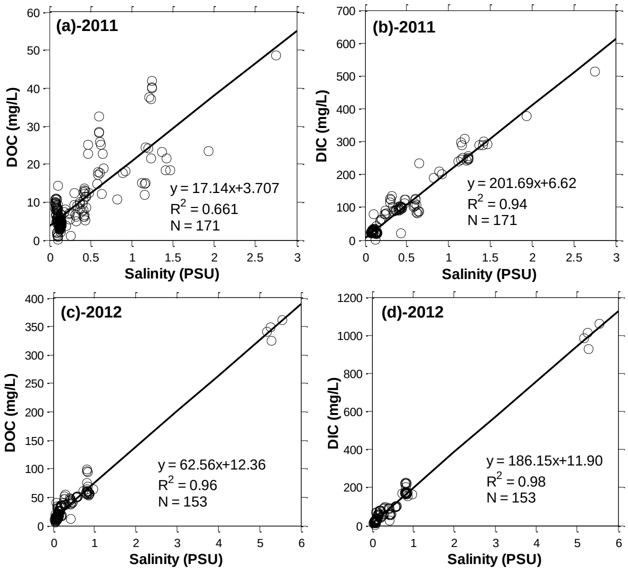

Fig. 6. Correlation between dissolved carbon and salinity (in PSU) in water bodies across the Songnen Plain. **(a)** DOC vs. salinity and **(b)** DIC vs. salinity for 2011 data set collected in late summer to early autumn; **(c)** DOC vs. salinity and **(d)** DIC vs. salinity with independent samples were collected in spring 2012 to test stability of the relationship between dissolved organic carbon and salinity.

mineralization of DOC in these waters (Granéli et al., 1996; Duarte et al., 2008). Land use may play an important role in the higher DIC levels in these terminal waters due to the saline–alkaline soil around these water bodies (Fig. 1b–c) (Wetzel 2001; Duarte et al., 2008; Wilson and Xenopoulos, 2008; Philips, 2010).

In this study, $SUVA_{254}$ is proven to be an effective index for characterizing DOC concentrations. The $SUVA_{254}$ reveals that high molecular weight DOC fractions are abundant in fresh waters due to quick exchange rates of DOC in the water column and less residence time (Spencer et al., 2008, 2012). Due to a long residence time for the brackish waters (Table 1), DOC concentrations increased, even though limited inflow led to decreased DOC transportation compared to humid or semi-humid regions (Duarte et al., 2008). These findings suggest that some DOC inputs can be evapo-concentrated (Curtis and Adams, 1995). Despite the prolonged photo-oxidation or the autochthonous DOC origins, the lower molecular weight DOC fraction is more abundant in brackish waters (Weishaar et al., 2003).

High values of both $E_{250:365}$ and Sr are observed in brackish waters, which may be regulated by the DOC flux for inland waters both in time and space (Jaffé et al., 2008; Spencer et al., 2009, 2012; Zhang et al., 2010; Fellman et al., 2011). Nevertheless, the $E_{250:365}$ and Sr values are found to be useful for indicating seasonal DOC compositional variations (Fichot and Benner, 2011). Relatively short residence times in fresh waters enhanced terrestrial DOC input, resulting in high molecular weight DOC fractions (Table 1). In contrast, long irradiation times in mid-west of the Songnen Plain generally decreased the high molecular weight fractions (Helms et al., 2008). Note that the special behavior of $SUVA_{254}$, $E_{250:365}$ and Sr in Nanhu Lake is likely due to the fact that the sewage water has different DOM composition, resulting in different CDOM characteristics compared to other fresh waters.

To analyze the climatic variables' influence on dissolved carbon in waters across the Songnen Plain further, multiple linear regression analyses were performed on both DOC and DIC using precipitation (P), potential evapotranspiration (ET_0, calculated based on FAO–Penman–Monteith equation), and sunshine duration hours (S) as independent variables. As expected, the ET_0 has large impacts on DOC with respect to precipitation and sunshine duration hours (see slope value for ET_0 in Table 4). The ET_0 representing the water output for the brackish waters results in the condensed effect on DOC concentration. In contrast, an inverse trend was observed between DIC and climatic variables (Table 4). The observation indicates that climatic variables exerted significant impacts on DOC and DIC concentration in the Songnen Plain. So far, our explanation is that sunshine duration may enhance photobleaching that could speed up DOC conversion to DIC (Wetzel, 2001; Lapierre et al., 2012). Further investigation is needed to examine possible underlying reasons.

4.2 Seasonal variation

The DOC in waters showed seasonal dynamics due to hydrological, climatic and landscape variation (Jaffé et al., 2008; Spencer et al., 2012). As mentioned above, high DOC concentrations in both the STR and CGL in May are attributed to winter accumulated dissolved carbon in soil flushed into stream or lakes during the snowmelt season in cold temperate zones (Ågren et al., 2007; C. C. Song et al., 2011). Similar findings have been reported in streams in Sweden (Ågren et al., 2007, 2010), in water across USA (Jaffé et al., 2008; Helms et al., 2008), and catchments that drain into the Arctic Ocean (Neff et al., 2006; Spencer et al., 2008). The DOC concentration in fresh waters in the ice-free season can be very variable, depending on the hydrological and climatic condition, and landscape in the catchment (Wetzel, 2001; Jaffé et al., 2008; Spencer et al., 2012). However, low DOC is generally expected in fresh waters during low flow season due to limited allochthonous input. Decreased autochthonous origin due to low algal abundance resulting from temperature in the STR could be another reason for the relatively low DOC concentration in October (Fig. 4a). The high DOC concentrations for the CGL in autumn are probably due to the fact that evaporation exceeds inflow and results in condensed DOC concentration. In summer (July, August and September), a large amount of recipient water from precipitation and runoff diluted DOC in the water is the main reason for the low DOC concentration (Fig. 4a).

High molecular weight DOC is more abundant in the early stages of the growing season (May) in the Songnen Plain, and large molecular weight CDOM prevails in the spring with snowmelt as reported by Ågren et al. (2007, 2010). Overall, the STR shows higher SUVA_{254} values than the CGL due to its more terrestrial DOM inputs and less residence time (Hood et al., 2003). Note that overall patterns show large molecular CDOM dominates in fresh waters, and different indices (e.g., the SUVA_{254}, $E_{250:365}$ and Sr) demonstrate variations for CDOM characterizing in different seasons for both fresh and brackish waters (Fig. 5a–c). The underlying reason is probably due to the seasonal variation and major sources of CDOM concentration (Spencer et al., 2012), particularly after punctuated discharge events coming from different landscapes, which are consistent with investigations by Wilson and Xenopoulos (2008, 2009).

4.3 Dissolved carbon vs. salinity

Within semi-humid and semi-arid regions, dissolved carbon is related to salinity, reflecting water residence times and dissolved matter accumulation (Duarte et al., 2008; Mattsson et al., 2009). This relationship implies that the source and sink patterns are similar among lakes within semi-arid regions (Cole et al., 1994; Duarte et al., 2008). The most likely explanation is that the most resilient DOM is evapo-concentrated in the semi-arid region. The relationship of DIC and salinity is more consistent (Fig. 6b and d), while the slope and intercept for the DOC and salinity regression model vary (Fig. 6a and c). Comparatively, DIC is more stable, which explains the stable relationship between DIC and salinity (Lapierre and Giorgio, 2012). An empirical model can be calibrated with a comprehensive data set covering waters across the Songnen Plain for estimating DIC storage. However, DOC is more labile (more sensitive to photochemical and microbial degradation) with different source (allochthonous or autochthonous) and composition (Spencer et al., 2009, 2012), resulting in different relationships between DOC and salinity. If a model is calibrated with data sets collected in various seasons, it is also possible to estimate DOC concentration in the Songnen Plain for the variance ($R^2 > 0.6$) it could explain.

4.4 Future implications

The ability to derive DIC concentration via salinity in the Songnen Plain implies improved carbon storage estimates in the semi-arid region. To quantify the carbon cycling for inland waters, calculation of the flux of dissolved carbon for inland waters is dependent on accurately constraining the concentration and volume of water bodies (Cole et al., 2007; Duarte et al., 2008; Tranvik et al., 2009). Thereby any technique that can improve flux estimates is of assistance in improving fresh and brackish waters delineation and DIC estimates. Recent studies have realized the limitation for quantifying carbon flux of inland brackish waters (Cole et al., 2007; Duarte et al., 2008; Tranvik et al., 2009). The correlation between DIC and salinity from the Songnen Plain strengthens the ability to increase spatiotemporal resolution easily through quick in situ data collection. If such an approach across other watersheds with similar conditions proves to be effective, it will lead to quick estimates of dissolved carbon storage coupled with remote sensing technology (Cole et al., 2007; Tranvik et al., 2009) for providing water surface area, and possible volume (Hendriks et al., 2012). However, this relationship generally varies depending on the source of CDOM and DOC (Spencer et al., 2009, 2012). Coupled with remote sensing, the close relationship between CDOM and DOC provides potential for dissolved carbon storage estimates (Griffin et al., 2011).

5 Conclusions

The knowledge of DOC and DIC in brackish inland waters is rare and incomplete. However, it has important implications for carbon cycling in saline (includes brackish) inland waters. A comprehensive study is require for understanding dissolved carbon characteristics that facilitate carbon cycling estimates for inland waters, particularly brackish waters in semi-arid or arid regions. For the first time, the characteristics of DOC and DIC in fresh and brackish waters in the Songnen Plain, China, have been investigated. This study provides insight into carbon cycles linked to hydro-climatic conditions in semi-arid inland waters. The following can be concluded: (1) compared to open waters in the southeast of the Songnen Plain, high DOC and DIC concentrations are observed in the brackish waters in the western part of the plain; (2) terminal aquatic environments result in elevated DOC and DIC concentration, and significant correlations between salinity–DOC and salinity–DIC providing a means for predicting dissolved carbon estimates from salinity; (3) relatively high molecular weight DOC fractions are more abundant in fresh waters. The elevated DOC and DIC levels in the semi-arid Songnen Plain might inspire further investigation for understanding the carbon cycling process in brackish aquatic ecosystems, which has not yet been fully done for saline (including brackish) inland waters across the world.

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