**RESEARCH ARTICLE** 



# **Constructed wetlands may lower inorganic nutrient inputs but enhance DOC loadings into a drinking water reservoir in North Wales**

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Received: 24 September 2015 / Accepted: 26 May 2016 / Published online: 4 June 2016 © Springer-Verlag Berlin Heidelberg 2016

**Abstract** The objective of this study was to monitor a newly constructed wetland (CW) in north Wales, UK, to assess whether it contributes to an improvement in water quality (nutrient removal) of a nearby drinking water reservoir. Inflow and outflow of the Free Water Surface (FWS) CW were monitored on a weekly basis and over a period of 6 months. Physicochemical parameters including pH, conductivity and dissolved oxygen (DO) were measured, as well as nutrients and dissolved organic and inorganic carbon (DOC, DIC) concentration. The CW was seen to contribute to water quality improvement; results show that nutrient removal took place within weeks after construction. It was found that 72 % of initial nitrate (N0<sub>3</sub><sup>-</sup>), 53 % of initial phosphate (PO<sub>4</sub><sup>3-</sup>) and 35 % of initial biological oxygen demand (BOD) were removed, calculated as a total over the whole sampling period. From our study, it can be concluded that while inorganic nutrients do decline in CWs, the DOC outputs increases. This may suggest that CWs represent a source for DOC. To assess the carbon in- and output a C budget was calculated.

Keywords Nutrient removal  $\cdot$  Carbon budget  $\cdot$  Constructed wetland  $\cdot$  Surface flow wetland  $\cdot$  DOC  $\cdot$  Drinking water treatment

Responsible editor: Philippe Garrigues

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#### Introduction

Wetlands function as sinks for nutrients and are therefore often used to improve water quality. The ability of wetlands to sequester nutrients is due to two processes; the uptake and immobilisation of nutrients by plant, microorganisms and soil matrix (Corbitt and Bowen 1994) and suppressed rates of decomposition of organic matter and re-release of nutrients in anaerobic wetland sediments (Clymo and Reddaway 1971). Therefore, CWs offer a natural solution for removing nutrient pollution in aquatic ecosystems. Such low-tech treatment systems are often more economically favourable than energy-intensive engineered treatment plants, are easier to operate and can provide numerous secondary benefits such as improved biodiversity. CWs are increasingly used to reduce concentrations of nitrate and phosphate concentrations in surface waters to prevent eutrophication (Kadlec 2012). In England and Wales, two-thirds of drinking water comes from surface water (reservoirs, lakes and rivers) while one-third is taken from groundwater. In addition to nutrients, surface waters can contain naturally high concentrations of DOC. The removal of DOC is a costly and energy-intensive stage of water treatment (Jones et al. 2015) as DOC compounds persisting until the disinfectant stage can react with the disinfectant (usually chlorine) to produce carcinogenic disinfection by-products (DBPs) (Chow et al. 2003). Another recent concern is the increasing amount of DOC in our rivers and streams. A study by Evans et al. (2005) estimated an increase in DOC in UK waters by 91 %. The worry about rising DOC concentration is due to effect it can have on surface water pH and acid neutralising capacity (Kullberg et al. 1993). In addition, high DOC concentrations reduce light penetration in water by giving it a brownish shade. This affects the aquatic productivity, which has implications for the aquatic food chain and lake stratification (Keller et al.

2003). High DOC concentrations in surface waters that feed reservoirs are linked to catchment properties, hydrological conditions, land management characteristics and climatic conditions (Pacheco et al. 2013). DOC originates from either autochthonous or allochthonous sources; the autochthonous organic carbon pool is mainly controlled by algae and plant production and the allochthonous by climate and processes within the catchment (Junlong et al. 1997). Eutrophication, which affects many reservoirs, may present a major problem as the resulting enhanced algal growths may also increase the input of DOC, thereby increasing the potential risk of DBP formation (Gough et al. 2015). Only a few studies about C budgets of CWs exist, and their results are variable but confirm that CWs often act as a net source for DOC (Kovacic et al. 2000, 2006). The objective of this study is to monitor a newly constructed wetland on a weekly interval for its physiochemical parameters at the inflow and outflow, to evaluate the water quality improvement. We hope to make a statement about how long it takes for a new CW to remove nutrients and evaluate the removal efficiency on a high resolution time span.

# Material and methods

#### Site description

The surface flow CW (Fig. 1) is located on one of the inflow streams to a drinking water reservoir in north Wales. It is a forested area, with mainly coniferous trees. The reservoir in this study is eutrophic due to agricultural practices within the catchment and the streams flowing into the reservoirs have moderate to high DOC concentrations (~10 mg/l). The area around the reservoir is used for cattle and sheep farming aided by modern agro-chemicals (Hughes et al. 2013). The bedrock at the site consists dominantly of Schist, and the aquifer has a limited yield for groundwater resources. The primary aim of the CW is the removal of nitrogen and phosphorus, to minimise the growth of algae in the reservoir and the formation of DBPs at the treatment plant. Phosphorus and nitrogen are high due to significant agricultural influence.

The CW system consists of 414  $m^2$  of treatment area in addition to 81  $m^2$  of open water. The nominal hydraulic retention time (nHRT) is 3 days. Average inflow flow rate is 1.02 L/s. The length of the system is 33 m, width at the inflow is 12 and 18 m at the outflow. Furthermore, 1.8 m was added as open water zone, which allows oxygenation, increased retention time and provides mixing, which can enhance removal processes. The treatment area comprises of a range of naturally occurring reeds, like phragmites. Only material from the site was used for construction of the walls and a series of baffles, and a plastic outflow pipe was added. The flow worked as a natural system through gravity, no additional pumping was used. In this study, we aim to investigate the time in which it takes a newly constructed wetland to improve and enhance the water quality of an inflow into a eutrophic drinking water reservoir. Therefore, physicochemical parameters at the inflow and outflow were measured on a weekly basis over a period of 6 months and a C budget was calculated.

#### Field and laboratory techniques

Sampling was undertaken every week between 27/03/2014 and 24/10/2014. Sampling of the CW outflow started 11 days later from the 07/04 onwards. Mean temperature over the sampling period was 13.4 °C and average rainfall was 3.0 mm/day. In situ measurements of temperature and DO (Milwaukee Instruments MW-600 Smart DO Meter) were made each time. At the inflow and outflow, three samples were collected using a 1-L glass bottle, a 100-ml plastic bottle and a 50-ml amber glass bottle (to minimise UV influences on samples). The 1-L glass bottle was used to measure particulate organic carbon (POC) by high temperature combustion (550 °C for 2 h) of the sample on a GF/F Whatman glass fibre filter. The 100-ml plastic bottle was completely filled and left unfiltered to measure pH (SevenEasy pH meter, Mettler Toldeo), conductivity (Orion 5 probe), bicarbonate and dissolved greenhouse gas concentrations. Bicarbonate was measured by taking 10 ml of unfiltered sample and titrating to pH 4.3 with 0.1 M HCl. Dissolved gases were determined using a similar headspace equilibrium method as that described by Dawson et al. (2002), with the gases analysed using a Varian 450 GC. The remaining sample was filtered (0.45 µm Whatman Glass-fibre filters) and analysed for DOC and DIC concentrations (Thermalox TC/TN, Analytical Sciences Ltd.), specific ultraviolet absorbance (SUVA) (Spectromax M2e Spectrophotometer, Molecular Devices) and nutrients (850 IC, Metrohm). The SUVA value gave information about the aromaticity of the water sample and is calculated with the DOC (mg/l) and the UV absorbance at 254 nm. The 50-ml amber bottle was incubated at room temperature for 5 days, so biological oxygen demand (BOD) could be calculated. To assess carbon processing, the following mass balance equation was applied (slightly altered from Pacheco et al. 2013):

$$POCin + DOCin + DICin + TOCdep$$
  
= POCout + DOCout + DICout + OCs + CO2  
+  $\Delta TCst$ 

where TOC is total organic carbon, OC is organic carbon, the subscripted "in" signifies the inflow from the catchment, "dep" is atmospheric wet deposition, "out" is outflow from the lake, "s" is permanent burial in sediment, and  $CO_2$  is the

# Fig. 1 Constructed wetland 3D schematic



concentration of  $CO_2$  in the water. Additionally,  $CH_4$  was calculated as a  $CO_2$  equivalent and added to the  $CO_2$ .  $\Delta TCst$  is the change in the pool of C in the lake over each sampling time period (1 week). POC<sub>in</sub>, DOC<sub>in</sub>, DIC<sub>in</sub>, POC<sub>out</sub>, DOC<sub>out</sub>, DIC<sub>out</sub> were calculated from weekly measured concentrations in streams.

To assess the effectiveness of the wetland nutrient removal efficiency (R) was calculated as follow

$$R(\%) = \left(\frac{Ci-Co}{Ci}\right) *100$$

Nutrient concentration influent (Ci) was measured at the inflow and the nutrient concentration effluent (Co) was sampled at the outflow both on a weekly basis and for calculation of R the average mean value was used. Pearson correlation factors for all parameters were calculated with SPSS.

# Results

#### **Dissolved and particulate carbon**

Mean DOC concentration of the inflow was  $7.22\pm0.17$  mg/l and at the outflow it was  $11.43\pm0.21$  mg/l, with the concentration of the outflow in general higher than the inflow and both exhibiting the same approximate seasonal trend (Fig. 2). DOC accumulation in the CW was 60 % as an average over the entire sampling period. DIC concentration at the inflow was  $18.07\pm0.21$  and  $16.24\pm0.37$  mg/l at the outflow. Removal of DIC was approximately 10 %. POC concentration at the inflow was  $0.82\pm0.04$  mg/l and  $2.00\pm0.09$  mg/l at the outflow. Pearson correlation coefficients for DIC, removal DIC, DOC, removal DOC, removal POC, SUVA and removal SUVA were calculated with SPSS (Table 1).

#### **SUVA**

Mean SUVA value at the inflow was  $3.49\pm0.06$  and for the outflow  $3.13\pm0.04$ . After initially high (>4) SUVA values at the outflow during start-up phase, SUVA concentration stabilises to values lower than four at the end of April. Outflow waters have a higher DOC concentration and higher UV<sub>254</sub> absorbance.

#### Nutrients

Mean nitrate concentrations were  $4.5\pm0.15$  mg/l (inflow) and  $1.2\pm0.09$  mg/l (outflow) (Fig. 3). Nitrate removal was measured 2 months after construction and the removal rate over the whole sampling period were 72 %. Nitrate removal rates were significantly lower in autumn, with an average of 30–



Fig. 2 Dissolved organic carbon (DOC) concentration (mg/l) at the inflow and outflow of the constructed wetland over the sampling period (April to October)

**Table 1**Carbon parametersPearson correlation

Correlation	DOC (mg/L)	R DOC (mg/L)	DIC (mg/L)	R DIC (%)	R POC (%)	SUVA	R SUVA
Pearson correlation	Br <sup>-</sup> -0.521** SO <sub>4</sub> <sup>-</sup> 0.530** HCO <sub>3</sub> 0.564** R SUVA 0.481*	R UV <sub>254</sub> 0.900** R SUVA -0.668** R NO <sub>3</sub> -0.462* R CH <sub>4</sub> 0.471*	0 <sub>2</sub> -0.736** Ca <sup>2+</sup> 0.805** HCO <sub>3</sub> 0.889** ANC 0.853** R NO <sub>3</sub> 0.676**	R UV <sub>254</sub> 0.900** R SUVA -0.668** R NO <sub>3</sub> -0.462* R CH <sub>4</sub> 0.471*	R CH <sub>4</sub> 0.454*	Mg <sup>2+</sup> 0 517** R DOC -0.581** R SUVA 0.643** R PO <sub>4</sub> 0.622*	R 0 <sub>2</sub> 0.490* R Br <sup>-</sup> -0.525* R PO <sub>4</sub> <sup>-</sup> 0.577*

\*\*Correlation is significant at the 0.01 level (2-tailed); \*correlation is significant at the 0.05 level (2-tailed)

40 %. MMR of nitrate was 0.70 g  $\cdot$  m<sup>-2</sup> day<sup>-1</sup>. Removal of DIC and O<sub>2</sub> was negatively correlated with NO<sub>3</sub> removal rates (Table 2).

Mean phosphate concentration was  $0.04 \pm 0.0018$  mg/l (inflow) and  $0.02 \pm 0.0021$  mg/l (outflow). Phosphate removal was 53.4 % as an average over the sampling period and positively correlated with Nitrate removal (Table 2). Phosphate MMR was 0.0043 g m<sup>-2</sup> day<sup>-1</sup>. Phosphate concentrations are much lower at the outflow than at the inflow (Fig. 4)

Mean bromide concentration (Fig. 5) at the inflow was  $0.07 \pm 0.0007$  and  $0.1 \pm 0.0012$  mg/l at the outflow. MMR was calculated as -0.0049 g m<sup>-2</sup> day<sup>-1</sup>. Nutrient removal efficiency was -31.5 %.

#### Dissolved oxygen, BOD<sub>5</sub>, temperature and pH

The average DO value at the inflow was  $8.26 \pm 0.12$  mg/l, and at the outflow, it was  $7.89 \pm 0.22$  mg/l. Dissolved oxygen



**Fig. 3** Bromide concentrations (mg/l) at the inflow (*blue*) and outflow (*red*) of the CW, plotted over the sampling period April–August

increased by 4.5 % from inflow to outflow, and the mean BOD<sub>5</sub> was reduced by 46 %. The average temperature at the inflow was  $12.99\pm0.08$  °C. Outflow temperature was  $19.5\pm0.2$  °C on average, and the wetland was by average  $6.6^{\circ}$  warmer than the inflow. Mean inflow pH was  $7.7\pm0.2$ , and at the outflow, it was  $7.6\pm0.3$ . By average, the pH was 0.1 lower at the outflow than at the outflow. Table 3 shows significant correlations between the discussed parameters.

#### **Greenhouse Gases**

The concentrations of dissolved greenhouse gases (GHGs), especially CH4, were substantially higher in the outflow compared to inflow (Table 4). Average CH<sub>4</sub> emission at the inflow was  $2.11 \pm 0.16 \mu g/l$ , and at the outflow, it was  $20.11 \pm 0.56 \mu g/l$ . CH<sub>4</sub> concentration was especially high in the summer when the water was warmer (Fig. 6). The CO<sub>2</sub> concentration was 737.89±20.94 mg/l (inflow) and 1128.67±25.15 mg/l (outflow). Removal efficiency for CO<sub>2</sub> was approximately -53 %. The outflow concentration increased steadily until the

Table 2 Nutrients Pearson correlation

Correlation	$NO_3^-$	R NO <sub>3</sub> <sup>-</sup>	$PO_4^{-}$	Br
Pearson correlation	CO <sub>2</sub> -0.462**	R POD <sub>4</sub> <sup>-</sup> 0.941**	CH <sub>4</sub> -0.524**	NO <sub>3</sub> 0.417**
	R DIC -0.455**	Temp 0.517*	CO <sub>2</sub> -0.559**	CH <sub>4</sub> 0.470**
	R O <sub>2</sub> -0.650**		R POC -0.557**	R Br <sup>-</sup> 0.443**
	R Br <sup>-</sup> 0.476** RCH <sub>4</sub> -0.456**		RCH <sub>4</sub> -0.575**	

\*\*Correlation is significant at the 0.01 level (2-tailed); \*correlation is significant at the 0.05 level (2-tailed)



**Fig. 4** Phosphate concentrations (mg/l) at the inflow (*blue*) and outflow (*red*) of the CW, plotted over the sampling period April–August

end of September; afterwards, it declined and dropped under the concentration of the inflow (Fig. 7).

#### **Carbon budget**

The carbon budget (Fig. 8) shows that, over the entire sampling period, 17.2 % less DIC, 11.5 % more DOC and approximately twice as much POC flowed into the reservoir compared to the amounts that are expected without the presence of the CW. Of the carbon, 7.5 % is retained in the wetland through sedimentation and plant uptake. Nutrient removal efficiency is -60.7 %. Mass removal rate (MRR) was -0.92 g m<sup>-2</sup> day<sup>-1</sup> calculated as mean average over the whole sampling period.

### Discussion

This study aimed to investigate the nutrient removal capability of a new CW, calculate its carbon budget and determine its impact on DOC loading to a reservoir.



Fig. 5 Nitrate concentrations (mg/l) at the inflow (*blue*) and outflow (*red*) of the CW, plotted over the sampling period April–August

Nitrate removal is expected to be controlled by three main mechanisms: (1) denitrification, (2) plant uptake and (3) mineralization (Vymazal 2007). The higher nitrate concentration during the startup phase is most likely due to the growth of the vegetation around the area and may partly be due to the wash off of nutrients during the construction phase. The CW functions as a nitrate source in autumn; this may be related to plant senescence and rapid mineralization of organic N within the system, while very high removal rates in summer can be explained by active denitrification. For denitrification, DOC is a key factor as well as DO, as an anaerobic environment is needed. As found in previous research, denitrification rates are mainly constrained by environmental conditions such as temperature, pH and carbon availability (Song et al. 2011 and Bachand and Horne 2000).

Initial phosphate concentration at the inflow most likely originates from  $PO_4^{3-}$  in the soil, which was released during construction work. Later, increasing phosphate concentrations were probably due to agricultural nutrients leaching into the surface water, which is beneficial for plant growth. According to Verhoeven and Meuleman (1999), phosphate removal in most CWs does not exceed more than 50 %. A review study by Vymazal (2007) stated total phosphate removal rates between 40 and 60 % and total nitrogen removal rates between 40 and 55 %, for various types of CWs. Therefore, the removal rates, phosphate 53 % and nitrate 73 %, achieved during the startup phase of this CW show a high performance quality.

While inorganic nutrients were sequestered in this CW, achieving the main aim of their installation, the DOC outputs increased. Since only one site was monitored during this study, broad conclusions about the impacts of CWs on DOC cannot be made, but the enhanced DOC loading to the reservoir is a significant water quality issue at this site because the reservoir as used as a source of drinking water. DOC can be leached into water flowing through wetlands as plants, algae, and bacteria grow, die and decay (Pinney et al. 2000; Villa et al. 2014). A study by Xenopoulos et al. (2003) investigated the amount of DOC loadings from different catchment areas; the results showed that forested wetlands, especially those with coniferous vegetation, have the highest leaching of DOC into the environment.

Particulate organic carbon causes the formation of anaerobic microsites, supporting simultaneous nitrification and denitrification. Therefore, it may play a dual role in denitrification, since it supports the heterotrophic metabolism of denitrifying bacteria as well as the  $O_2$  consumption which creates anaerobic microsites necessary for denitrification (Hamersley and Howes 2002). The accumulation of twice as much POC in the constructed wetland enhances denitrification processes. Bioavailable POC acts not only as a C substrate for denitrifiers, but also depletes DO levels within particles via aerobic respiration, supporting denitrification within aerobic wastewaters. According to Moran and Hodson (1994), higher POC

Table 3	Environmental	parameters	Pearson	correlation
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Correlation	DO (mg/L)	BOD (mg/L)	рН	R pH	Temperature (°C)	Conductivity (µ g/L)
Pearson correlation	BOD 0.822**	Cl <sup>-</sup> 0.696**	R NO <sub>3</sub> 0.425*	R O <sub>2</sub> -0.636**	Cond. 0.578**	SUVA 0.537**
	Cl <sup>-</sup> 0.586**	CH <sub>4</sub> 0.646**		R CO <sub>2</sub> 0.572**	DIC 0.591**	Cl- 0.518**
	HCO <sub>3</sub> 0.754**	ANC 0.595**				DIC 0.498*
	ANC 0.972**	R UV <sub>254</sub> 0.616**				SO <sub>4</sub> <sup>-</sup> 0.399*

\*\*Correlation is significant at the 0.01 level (2-tailed); \*correlation is significant at the 0.05 level (2-tailed)

additions results in higher respiration rates, and the faster creation of anaerobic microsite volume to support denitrification.

According to Lin et al. (2002), macrophytes present species-specific nitrate removal efficiency, depending on their ability to produce carbon for denitrification. DOC and bromide accumulation increases the likelihood of DB production in the form of trihalomthanes (THMs) and haloacetonitriles (HANs) during water treatment. Some of these DBPs are suspected to be mutagens, carcinogens or developmental toxicants if ingested over extended periods of time (Villanueva et al. 2004). Bromide is a major inorganic DBP precursor that results in formation of brominated DBPs, which are generally more toxic than chlorinated DBPs (Richardson et al. 2007). According to Ingersoll and Baker (1998), when nitrate removal efficiencies increase, dissolved organic carbon in the effluent also increases, as does chloroform formation potential. Bacterial decomposition of plant detritus has been shown to convert POC into dissolved form and cause the release of humic substances into the bulk DOC pool (Moran and Hodson 1994).

SUVA is a reliable indicator of DOC aromaticity and liability of natural organic matter (NOM) to coagulation, and it is known that the NOM in high-SUVA waters tends to have lower alkalinities and hardness and higher TOC concentrations and is therefore more amenable to removal by coagulation at the treatment works (Archer and Singer 2006). Results

Table 4 Greenhouse gases (GHG) Pearson correlation

Correlation	CH4	R CH4	CO
	0114	IC CI14	002
Pearson correlation	CO <sub>2</sub> 0.742**	R CO <sub>2</sub> 0.561 **	R CH <sub>4</sub> 0.595**
	R UV <sub>254</sub> 0.484*		R CO <sub>2</sub> 0.440*
	R CH <sub>4</sub> 0.457*		

\*\*Correlation is significant at the 0.01 level (2-tailed); \*correlation is significant at the 0.05 level (2-tailed)

show that despite the rise in DOC concentration at the outflow, SUVA values decrease. This indicates the added DOC will be harder to treat, but the CW will ultimately reduce algae in the reservoir, which will lead to higher SUVA values (Nguyen et al. 2005) and easier to treat water. The rather high (>4) SUVA values, during the first few weeks, could be due to disturbance effect of the CW construction.

Dissolved oxygen increased through the CW, probably due to the growing vegetation in the startup phase. A survey conducted by Puigagut et al. (2007) stated that BOD<sub>5</sub> removal generally ranged from 80 to 95 %. Even though BOD removal in the studied CW has a much lower removal rate, we assume that much oxygen was added through the free water area and cannot find a negative impact on other removal parameters. Kadlec and Wallace (2009) stated that BOD removal rates in CWs are always lower in summer.

GHGs were monitored to assess the carbon budget and to provide additional insight into the CW functionality. The concentration pattern of GHGs can be explained by vegetation growth phases over the sampling period. GHG concentration in CWs can vary significantly, depending on the kind of



**Fig. 6** CH<sub>4</sub> concentrations ( $\mu$ g/l) at the inflow (*blue*) and outflow (*red*) of the CW, plotted over the sampling period April–August



**Fig.** 7 CO<sub>2</sub> concentrations (mg/l) at the inflow (*blue*) and outflow (*red*) of the CW, plotted over the sampling period April–August

wetland, its location and various other parameters (Kang and Freeman 2002). CH<sub>4</sub> concentration showed high concentrations over spring; during the startup phase and a decline over the summer months and an increase during autumn, these results are the similar with the findings of Kang and Freeman 2002. The study suggests that higher CH<sub>4</sub> emissions from swamps in autumn were linked with litterfall production, which indicates a coupling between carbon input and methanogenesis. We found a positive correlation between DOC and CH<sub>4</sub> increases (Table 1). The high CH<sub>4</sub> concentration over spring in the outflow is mostly likely due to the rapidly growing vegetation around the wetland and the leaching of DOC from plants. Plants are the main source of carbon for microorganisms in CWs. This carbon is further transformed to gaseous forms and increases the loading of CO<sub>2</sub> and CH<sub>4</sub> into the wetland. Due to increased litterfall

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during autumn, the high  $CH_4$  and  $CO_2$  concentrations in September can be explained. Furthermore, plants increase the efficiency of nitrogen removal by supporting denitrifying microorganisms with easily decomposable organic matter (Picek et al. 2007). In a study conducted by Liikanen et al. (2006), it was estimated that even if all global wastewaters were treated in constructed wetlands, their share in atmospheric liability would be less than 1 % in total.

## Conclusion

As newly CWs have not been previously monitored during startup phase, weekly monitoring over a 6-month period was meant to give insight into the early stage functionality of a new CW. This study showed that newly constructed wetlands contribute to water quality improvement within a few weeks after construction. Nitrate and BOD<sub>5</sub> removal started showing an effect within 4 weeks after construction. Nitrate removal was the fastest and most efficient process, probably boosted by the very rapidly establishing vegetation around the CW. The early denitrification process could also be initiated by the carbon content of the soil or water. The plants leached organic matter into the wetland, which leads to enhanced DOC loading to the reservoir. By the beginning of September, nitrate had accumulated in the CW, possibly because vegetation started to decrease and could not take up nitrate as efficiently as over the spring/ summer months. Evergreen plants for vegetation might be a valid solution to produce less organic matter and take up nitrate all year around. Overall, spring was a good time to set up the CW, since natural vegetation developed fastest and promoted the CW's productivity.



Fig. 8 Carbon budget of CW, mean average concentration over the whole sampling period (April–October) Acknowledgments The authors thank the Wolfson Carbon Capture Laboratory Team for their support and the Natural Resources Wales and the MET office for providing data.

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