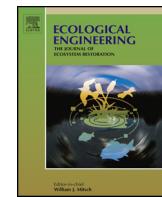




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# Greenhouse gas emission in constructed wetlands for wastewater treatment: A review

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## ABSTRACT

A literature analysis of 158 papers published in international peer-reviewed journals indexed by the Thomson Reuters Web of Knowledge from 1994 to 2013 showed that CO<sub>2</sub>–C emission was significantly lower in free water surface (FWS) constructed wetlands (CW) than in subsurface flow (SF) CWs (median values from 95.8 to 137.0 mg m<sup>-2</sup> h<sup>-1</sup>, respectively). In vertical subsurface flow (VSSF) CWs the CH<sub>4</sub>–C emission was significantly lower than in horizontal subsurface flow (HSSF) CWs (median values 3.0, 6.4, and 4.0 mg m<sup>-2</sup> h<sup>-1</sup>, respectively). There were no significant differences in N<sub>2</sub>O–N emission in various CW types (median for FWS, VSSF and HSSF CWs: 0.09, 0.12, and 0.13 mg m<sup>-2</sup> h<sup>-1</sup> correspondingly).

The highest value of emission factor (EF) of CH<sub>4</sub> ((CH<sub>4</sub>–C/inflow TOC<sub>in</sub>) \* 100%) was found for FWS CWs (median 18.0%), followed by HSSF CWs (3.8%), and VSSF CWs (1.28%). Median values of N<sub>2</sub>O EFs ((N<sub>2</sub>O–N/inflow TN<sub>in</sub>) \* 100%) differed significantly in all three CW types: 0.34% for HSSF, 0.11% for FWS, and 0.018% for VSSF CWs.

We found a significant correlation between TOC<sub>in</sub> and CH<sub>4</sub>–C emission and between the TN<sub>in</sub> and N<sub>2</sub>O–N emission values for all of the types of CWs we studied.

Hybrid CWs (e.g., the subsequent combination of VSSF, HSSF and FWS CWs) are beneficial from the point of view of both water purification and minimization of greenhouse gas (GHG) emissions. Likewise, intermittent loading in VSSF CWs and macrophyte harvesting in HSSF and FWS CWs can mitigate GHG emissions.

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## 1. Introduction

Constructed wetlands (CW) are engineered wetland systems that have been designed and constructed to utilize natural processes in treating wastewater (Vymazal et al., 1998). Constructed

wetlands are used to improve the quality of wastewater from point and nonpoint sources of water pollution, including domestic, industrial and municipal wastewater, stormwater runoff, farm wastewater, collated runoff from agricultural land and landfill leachate (Kadlec and Knight, 1996; Kadlec and Wallace, 2008).

The main types of CWs are: free water surface (FWS) or surface flow, vertical subsurface flow (VSSF) and horizontal subsurface flow (HSSF) CWs (Vymazal, 2007, 2011). In addition to wastewater treatment, the CWs provide several ecosystem services such as provisional (food, energy, fibers), regulating (carbon (C) sequestration, climate regulating, flood control), supporting (biodiversity,

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nutrient cycling) and cultural (recreational, educational) services (Mitsch and Gosselink, 2007).

*Free water surface CWs* are shallow and low flow velocity wetlands which have areas of open water and floating, submerged and/or emergent plants (Kadlec and Wallace, 2008). FWS CWs are very effective in the removal of organics through microbial degradation and the removal of suspended solids through filtration and sedimentation (Vymazal et al., 1998). The removal of nitrogen (N) and phosphorus (P) can be sustainable, but depends on inflow concentration, the chemical form of nitrogen, water temperature, the season, organic carbon availability, substrate material and dissolved oxygen concentration (Vymazal, 2011). The FWS wetlands are mostly used for the tertiary treatment of domestic and municipal wastewater, mine drainage waters, and for stormwater and agricultural runoff (Kadlec and Knight, 1996; Kadlec and Wallace, 2008).

In *horizontal subsurface flow CWs*, the wastewater is fed in at the inlet and flows slowly through the porous medium under the surface of the bed planted with emergent vegetation to the outlet, where it is collected before leaving via a water level control structure (Vymazal et al., 1998). During passage the wastewater comes into contact with a network of aerobic, anoxic and anaerobic zones. Most of the bed is anoxic/anaerobic due to the permanent saturation of the beds. The aerobic zones occur around roots and rhizomes that leak oxygen into the substrate (Brix, 1987). The most important properties of macrophytes planted in HSSF CWs are filtration bed insulation during the winter, substrate for the growth of attached bacteria, oxygen release to the rhizosphere, nutrient uptake and storage, C sequestration and root exudates with antimicrobial properties (Brix, 1997; Vymazal and Kröpfelova, 2008). HSSF CWs are commonly sealed with a liner to prevent seepage and to ensure controllable outflow, and are mostly used for secondary treatment of domestic and municipal wastewater (Vymazal and Kröpfelova, 2008). Organic compounds are degraded by bacteria under aerobic and anaerobic conditions. It has been shown that the oxygen transport capacity in these systems is insufficient to ensure aerobic decomposition and that anaerobic processes play an important role in HSSF CWs (Vymazal and Kröpfelova, 2008). Suspended solids settle into micropockets in the filtration bed or are filtered out. Removal of ammonia-N is limited by the lack of oxygen and hence nitrification in the filtration media. The HSSF CWs do, however, provide suitable conditions for denitrification (Vymazal and Kröpfelova, 2008). Removal of P is usually low unless special media with high sorption capacity are used. The selection of filtration material is also very important for the longevity of the system, because media that are too fine will clog the system, and surface runoff will occur (Vohla et al., 2011).

*Vertical subsurface flow CWs* comprise a flat bed of graded gravel topped with sand or other porous filter materials planted with macrophytes. In contrast to HSSF CWs, VSSF CWs are fed intermittently with large batches, thus flooding the surface. Wastewater then percolates down through the bed and is collected by a drainage network at the bottom. The bed drains completely, which allows air to refill the bed. The VSSF CWs provide greater oxygen transfer into the bed, thus producing a nitrified (high  $\text{NO}_3^-$ ) effluent (Cooper et al., 1996; Cooper, 2005). On the other hand, VSSF CWs do not provide suitable conditions for denitrification to complete conversion to gaseous nitrogen forms which escape to the atmosphere. Removal of organics and suspended solids is high (Vymazal and Kröpfelova, 2008). As compared to HSSF CWs, which need 5–6  $\text{m}^2$  per population equivalent (PE), vertical flow systems require less land, usually 1–3  $\text{m}^2 \text{ PE}^{-1}$  (Cooper, 2005).

Both VSSF and HSSF CWs with the ability to insulate the surface of the bed are capable of operation under colder conditions than are FWS systems (Mander and Janssen, 2003).

Various types of CWs are usually combined (i.e., hybrid or combined systems) in order to achieve higher removal efficiency, especially for nitrogen. The design commonly consists of two stages: several parallel VSSF beds followed by two or three HSSF beds in series (Vymazal, 2007). The VSSF wetland is intended to remove organics and suspended solids and to provide nitrification, while denitrification and the further removal of organics and suspended solids occur in the HSSF wetland. When aquatic macrophyte production is the main practical function of a wetland system, the VSSF–HSSF bed complex can be followed by a larger FWS wetland (Maddison et al., 2009).

As a bias of the water purification, the CWs for wastewater treatment have been found to be sources of greenhouse gases (GHG). Carbon dioxide ( $\text{CO}_2$ ) emission has been measured in few full-scale CWs (Mander et al., 2003, 2005a,b, 2008; Teiter and Mander, 2005; Liikanen et al., 2006; Ström et al., 2006; Garcia et al., 2007; Picek et al., 2007; Van der Zaag et al., 2010), and C balance has been compiled in only one HSSF CW based on the long-term direct measurement of C in inflow and outflow, accumulation in filter material (sand), microbes, above ground and below ground plant biomass, and the emission of  $\text{CO}_2$  and  $\text{CH}_4$  (Mander et al., 2008). On the other hand, there are more measurements of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emission from full-scale CWs:  $\text{CH}_4$  by Tanner et al. (1997), Xue et al. (1999), Johansson et al. (2004), and Chiemchaisri et al. (2008);  $\text{N}_2\text{O}$  by Fey et al. (1999) and Johansson et al. (2003); and both  $\text{CH}_4$  and  $\text{N}_2\text{O}$  by Tai et al. (2002), Wild et al. (2001), Mander et al. (2003, 2005a,b, 2008, 2011), Stadmark and Leonardson (2005), Teiter and Mander (2005), Liikanen et al. (2006), Søvik et al. (2006), Gui et al. (2007), Picek et al. (2007), Søvik and Kløve (2007), Ström et al. (2006), Liu et al. (2009), and Van der Zaag et al. (2010).

Recent research has shown that  $\text{N}_2\text{O}$  can be produced through a number of different pathways, both chemical and biochemical, during nitrification (stepwise conversion of ammonia to nitrate) and denitrification (stepwise conversion of nitrate to nitrogen gas; Colliver and Stephenson, 2000). Under aerobic conditions in a nitrifying wastewater treatment system,  $\text{N}_2\text{O}$  production through nitrifier denitrification has been identified as the predominant production pathway (Wunderlin et al., 2013; Aboobakar et al., 2013). Similarly, research from soil science has shown that in well-aerated, moist conditions (soil water filled pore space at 40–60%),  $\text{N}_2\text{O}$  can be emitted during nitrification (Robertson and Tiedje, 1987; Mosier, 1998; Mosier et al., 1998) by ammonia-oxidizing bacteria during the oxidation of hydroxylamine ( $\text{NH}_2\text{OH}$ ) to nitrite ( $\text{NO}_2^-$ ) (Arp and Stein, 2003), and also via reducing  $\text{NO}_2^-$  to  $\text{N}_2\text{O}$  and  $\text{N}_2$  under aerobic conditions by nitrifier denitrification (Goreau et al., 1980; Wrage et al., 2001).

Denitrification, as the microbial reduction of  $\text{NO}_3^-$ –N to  $\text{NO}_2^-$ –N and further to gaseous forms of NO,  $\text{N}_2\text{O}$  and  $\text{N}_2$  (Knowles, 1982), has been found in numerous studies to be a significant process in nitrogen removal in treatment wetlands (Bachand and Horne, 2000a,b; Spieles and Mitsch, 2000; Hernandez and Mitsch, 2006, 2007; Batson et al., 2012). Denitrification rates in soils are influenced by nitrate availability, carbon availability, temperature and pH (Mitsch and Gosselink, 2007). The last step of denitrification, i.e., the conversion of  $\text{N}_2\text{O}$  to  $\text{N}_2$ , is very sensitive to oxygen and redox status, and disruption of this step results in incomplete denitrification and  $\text{N}_2\text{O}$  emissions (Colliver and Stephenson, 2000). The relative contribution to  $\text{N}_2\text{O}$  emissions from a treatment system will depend on the environmental conditions that are generated and maintained throughout the pollutant transformation processes. Both denitrification and methane formation depend on the oxygen and redox status of the soil or sediment, which changes in both spatial and temporal contexts. In this relation, the variability of fluxes of both  $\text{N}_2\text{O}$  and  $\text{CH}_4$  is high (Willison et al., 1998; Teiter and Mander, 2005).

**Table 1**

Methane ( $\text{CH}_4-\text{C}$ ) and carbon dioxide ( $\text{CO}_2-\text{C}$ ) emission; inflow BOD or TOC values; inflow TOC loading ( $\text{TOC}_{\text{in}}$ ); and emission factor ( $\text{CH}_4-\text{C}/\text{TOC}_{\text{in}}$ ) in free water surface (FWS) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm, sTr – subtropical, n.a. – not available.

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q$ ( $\text{m}^{-3} \text{d}^{-1}$ )	BOD ( $\text{mg L}^{-1}$ )	TOC ( $\text{mg L}^{-1}$ )	Inflow BOD load ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{TOC}_{\text{in}}$ ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CO}_2-\text{C}$ emiss. ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4-\text{C}$ flux ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4-\text{C}/\text{TOC}_{\text{in}}$ (%)	References
Domestic wastewater	Nykvarn, Sweden	T	<i>Typha latifolia</i>	May–August 1998 April–October 1999	0.1	180	2.8	n.a.	21.0	10.5	n.a.	1.9	18	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Nykvarn, Sweden	T	<i>Phalaris arundinacea</i>	May–August 1998 April–October 1999	0.1	180	5.2	n.a.	39.0	19.5	n.a.	3.6	19	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Nykvarn, Sweden	T	<i>Glyceria maxima</i>	May–August 1998 April–October 1999	0.09	180	2.2	n.a.	18.3	9.2	n.a.	1.8	20	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Nykvarn, Sweden	T	<i>Lemna minor</i>	May–August 1998 April–October 1999	0.09	180	8.2	n.a.	68.3	34.2	n.a.	7.7	23	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Nykvarn, Sweden	T	<i>Spirogyra</i> spp.	May–August 1998 April–October 1999	0.09	180	2.7	n.a.	22.5	11.3	n.a.	1.9	17	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Nykvarn, Sweden	T	Plots without plants	May–August 1998 April–October 1999	0.09	180	3.3	n.a.	27.5	13.8	n.a.	2.8	20	Johansson et al. (2004) and Tonderski and Hansson (2001) for BOD data
Domestic wastewater	Lakeus, Finland	B	<i>Phragmites australis</i> , <i>T. latifolia</i>	January 2002–October 2003	4.4	3624	n.a.	25	n.a.	85.8	108.3	8.4	10	Søvik et al. (2006)
Domestic wastewater	Ruka, Finland	B	<i>Carex–Sphagnum</i>	January–October 2002	0.82	289	n.a.	17	n.a.	25.0	95.8	4.4	18	Søvik et al. (2006)
Domestic wastewater	Skallstuggu, Norway	B	<i>P. australis</i>	July 2001–February 2002	0.0065	n.a.	n.a.	n.a.	n.a.	22.0	87.5	5.8	26	Søvik et al. (2006)
Domestic wastewater	Skjønhaug, Norway	B	<i>Iris pseudacorus</i> , <i>T. latifolia</i> , <i>Schoenoplectus lacustris</i>	July 2001–February 2002	0.4	600	n.a.	26.7	n.a.	166.9	49.6	12.4	7	Søvik et al. (2006)
Domestic wastewater	Magle, Hässleholm, Sweden	T	<i>Juncus effusus</i> , <i>T. latifolia</i> , <i>P. australis</i>	April–May 2005	20	13,000	19	n.a.	51.5	25.7	100	4.8	19	Ström et al. (2006); <a href="http://www.hassleholmsvatten.se">www.hassleholmsvatten.se</a> for BOD data
Domestic wastewater	Skjønhaug, Norway	T	<i>I. pseudacorus</i> , <i>T. latifolia</i> , <i>S. lacustris</i>	October 2001–July 2002	0.4	520	15	n.a.	81.3	40.6	n.a.	181.0 <sup>a</sup>	446 <sup>a</sup>	Søvik and Kløve (2007)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	August 2004–May 2006	0.0012	0.5	107.3	10.3	n.a.	17.9	n.a.	4.6	26	Gui et al. (2007)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	June–August 2006	0.0012	0.5	200	n.a.	347.2	173.6	n.a.	27	16	Liu et al. (2009)
Agricult. non-point pollution	Donau-moos, Germany	T	Basin 1: <i>T. latifolia</i> , <i>T. angustifolia</i>	September 1998–May 1999	2.2	158	n.a.	3.5	n.a.	1.0	n.a.	0.15	14	Wild et al. (2001)
Agricult. non-point pollution	Donau-moos, Germany	T	Basin 2, <i>T. latifolia</i> , <i>T. angustifolia</i>	September 1998–May 1999	2.6	158	n.a.	11.2	n.a.	2.8	n.a.	0.88	31	Wild et al. (2001)
Agricult. non-point pollution	Genarp, Sweden	T	<i>Ceratophyllum demersum</i>	June 2003–May 2004	1	4286	n.a.	1.5	n.a.	26.8	n.a.	2.1	8	Stadmark and Leonardsson (2005); Swedish River Inventory for TOC data
Agricult. non-point pollution	Görarp, Sweden	T	No plants in plots	June 2003–May 2004	1.5	15,000	n.a.	1.3	n.a.	54.2	n.a.	2.6	5	Stadmark and Leonardsson (2005); Swedish River Inventory for TOC data
Agricult. non-point pollution	Ormastorp, Sweden	T	<i>L. minor</i>	June 2003–May 2004	0.7	3500	n.a.	1.8	n.a.	37.5	n.a.	3.1	8	Stadmark and Leonardsson (2005); Swedish River Inventory for TOC data

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q\text{ (m}^{-3}\text{ d}^{-1}\text{)}$	BOD ( $\text{mg L}^{-1}$ )	TOC ( $\text{mg L}^{-1}$ )	Inflow BOD load ( $\text{mg m}^{-2}\text{ h}^{-1}$ )	TOC <sub>in</sub> ( $\text{mg m}^{-2}\text{ h}^{-1}$ )	$\text{CO}_2\text{--C emiss.}$ ( $\text{mg m}^{-2}\text{ h}^{-1}$ )	$\text{CH}_4\text{--C flux}$ ( $\text{mg m}^{-2}\text{ h}^{-1}$ )	$\text{CH}_4\text{--C/TOC}_{\text{in}}$ (%)	References
Agricultural non-point pollution	Hovi, Finland	B	<i>T. latifolia</i> , <i>Scirpus sylvaticus</i> , <i>Alisma plantago-aquatica</i> , <i>P. arundinacea</i> , <i>Filipendula ulmaria</i> , <i>Iris pseudacorus</i> , <i>Juncus conglomeratus</i> , <i>Schoenoplectus validus</i>	January 2001–October 2002	0.6	78.6	n.a.	13	n.a.	41.9	29.4	1.6	4	Søvik et al. (2006)
Dairy farm wastewater	Ngatai, New Zealand	T/STr	<i>T. latifolia</i>	1994–1996	0.4	6.4	n.a.	n.a.	n.a.	57	n.a.	8.5	15	Tanner et al. (1997)
Dairy farm wastewater	Ngatai, Zealand	T/STr	<i>T. latifolia</i>	1994–1996	0.5	6.4	n.a.	n.a.	n.a.	57	n.a.	12.5	22	Tanner et al. (1997)
Dairy farm wastewater	Turro, Nova Scotia, Canada	T	<i>P. australis</i> , <i>Acorus calamus L. minor</i>	August 2005–September 2006	0.0007	0.105	92.3	n.a.	61.2	30.6	176	10.8	35	Van der Zaag et al. (2010)
Raw municipal wastewater	Jiaonan, China	T	<i>P. australis</i> , <i>Acorus calamus L. minor</i>	June 1999–August 2000	49.5	12,000	460	n.a.	464.6	232.3	n.a.	5220.0 <sup>a</sup>	2247 <sup>a</sup>	Tai et al. (2002)

<sup>a</sup> Not included in all calculations (see text).

Methane is produced in anoxic soils and sediments, while well-drained soils act as a sink for atmospheric CH<sub>4</sub> due to methane oxidation, through either ammonia oxidizers or methanotrophs (Le Mer and Roger, 2001). In treatment systems, such as wetlands, methane is generated from the anaerobic degradation of organic matter either present in the influent to the systems or accumulated as a result of plant litter accumulation. Methane generation occurs in the oxidation–reduction state range of –250 to –350 mV (Mitsch and Gosselink, 2007), in the absence of oxygen. In some conditions, it can start already at –150 mV (Wang et al., 1993). As such, emissions of methane from treatment wetlands are expected to be higher in anaerobic systems such as HSSF systems and, to a lesser extent, FWS and organically overloaded systems.

Due to increasing human impact on the global environment, nitrous oxide, which has an atmospheric lifetime of about 120 years, a global warming potential of 296 relative to CO<sub>2</sub> over a 100-year time horizon, and is responsible for about 6% of anticipated warming (IPCC, 2007), is increasing in the atmosphere at a rate of about 0.3% yr<sup>−1</sup>. Methane in the atmosphere has a lifetime of 8.4 years on a 100-year time horizon, CH<sub>4</sub> has a global warming potential of 25 relative to CO<sub>2</sub>, and is responsible for about 20% of anticipated warming (IPCC, 2007).

The main objectives of the study are: (1) to give an overview of GHG emissions and the main influencing factors in all types of CWs; (2) to analyze the relationship between CH<sub>4</sub> and N<sub>2</sub>O emission and related C and N loading in the inflow of CW systems; (3) to determine the emission factor (EF) values of CH<sub>4</sub> and N<sub>2</sub>O for different types of CWs.

Recently published reviews on N<sub>2</sub>O emission from CWs (Huang et al., 2013) and on climate regulation by FWS and created riverine wetlands (Mander et al., 2014) cover only part of CWs and a fraction of the available literature sources, whereas this paper seeks to cover the entire topic.

## 2. Materials and methods

### 2.1. Data sources and analysis

We reviewed 158 papers published in international peer-reviewed journals indexed by the Thomson Reuters Web of Knowledge from 1994 to 2013. The terms “free water surface”, “surface flow”, constructed wetland(s)”, “artificial wetland(s)”, “treatment wetland(s)”, “subsurface flow wetland(s)”, “vertical subsurface flow” and “horizontal subsurface flow” in combination with the terms “carbon dioxide”, “CO<sub>2</sub>”, “methane”, “CH<sub>4</sub>”, “nitrous oxide” and “N<sub>2</sub>O” were searched.

We found a total of 14 publications that provided information on emissions of either CH<sub>4</sub> or N<sub>2</sub>O ( $\text{g m}^{-2}\text{ h}^{-1}$ ) or both gases in FWS CWs. These publications presented information on 19 different SF CW systems, whereas for CH<sub>4</sub> and N<sub>2</sub>O there were 25 and 26 subsystems/measuring events, respectively, from which EF values could be calculated (Tables 1–6).

Regarding the vertical subsurface flow (VSSF) CWs, there were only 4 measurement periods presented for 3 CWs from which CH<sub>4</sub> emission data and EFs could be calculated: Kõo in Estonia (Teiter and Mander, 2005; Søvik et al., 2006), Ski in Norway (Søvik et al., 2006), and Miho/Ibaraki, Japan (Gui et al., 2007; Liu et al., 2009; Table 3). For N<sub>2</sub>O emissions, laboratory microcosm experiments with different plant species from Ibaraki, Japan (Inamori et al., 2008; Wang et al., 2008a,b) were included (Table 4).

Regarding CH<sub>4</sub> fluxes from HSSF CWs (Table 5) we possessed data from 9 systems. For N<sub>2</sub>O emissions from HSSFs, a CW for dairy

**Table 2**

Nitrous oxide ( $\text{N}_2\text{O}$ —N) emission; inflow total nitrogen loading ( $\text{TN}_{\text{in}}$ ); and emission factor ( $\text{N}_2\text{O}$ —N/ $\text{TN}_{\text{in}}$ ) in free water surface (FWS) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm, n.a. – not available.

Wastewater type	Site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q(\text{m}^{-3} \text{d}^{-1})$	$\text{TN}(\text{mg L}^{-1})$	$\text{TN}_{\text{in}}(\text{mg m}^{-2} \text{h}^{-1})$	$\text{N}_2\text{O}$ flux ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{N}_2\text{O}$ —N/ $\text{TN}_{\text{in}}$ (%)	References
Domestic wastewater	Nykvarn, Sweden	T	<i>T. latifolia</i>	May–August 1998 April–October 1999	0.1	180	8	60	0.081	0.14	Johansson et al. (2003)
Domestic wastewater	Nykvarn, Sweden	T	<i>P. arundinacea</i>	May–August 1998 April–October 1999	0.1	180	8	60	0.152	0.25	Johansson et al. (2003)
Domestic wastewater	Nykvarn, Sweden	T	<i>Glyceria maxima</i>	May–August 1998 April–October 1999	0.09	180	8	67	0.031	0.05	Johansson et al. (2003)
Domestic wastewater	Nykvarn, Sweden	T	<i>L. minor</i>	May–August 1998 April–October 1999	0.09	180	8	67	0.094	0.14	Johansson et al. (2003)
Domestic wastewater	Nykvarn, Sweden	T	<i>Spirogyra</i> spp.	May–August 1998 April–October 1999	0.09	180	8	67	0.036	0.05	Johansson et al. (2003)
Domestic wastewater	Nykvarn, Sweden	T	Plots without plants	May–August 1998 April–October 1999	0.09	180	8	67	0.192	0.29	Johansson et al. (2003)
Domestic wastewater	Lakeus, Finland	B	<i>P. australis</i> , <i>T. latifolia</i>	January 2002–October 2003	44	3624	66.1	23	0.007	0.03	Søvik et al. (2006)
Domestic wastewater	Ruka, Finland	B	<i>Carex</i> – <i>Sphagnum</i>	January–October 2002	0.82	289	59.7	88	0.106	0.12	Søvik et al. (2006)
Domestic wastewater	Skallstuggu, Norway	T	<i>P. australis</i>	July 2001–February 2002	n.a.	n.a.	n.a.	69.2	0.041	0.006	Søvik et al. (2006)
Domestic wastewater	Skjønhaug, Norway	T	<i>I. pseudacorus</i> , <i>T. latifolia</i> , <i>S. lacustris</i>	July 2001–February 2002	0.4	300	43.4	136	0.094	0.07	Søvik et al. (2006)
Domestic wastewater	Magle, Håssleholm, Sweden	T	<i>J. effusus</i> , <i>T. latifolia</i> , <i>P. australis</i>	April–May 2005	20	13,000	75	203	0.230	0.11	Ström et al. (2006)
Domestic wastewater	Skjønhaug, Norway	T	<i>I. pseudacorus</i> , <i>T. latifolia</i> , <i>S. lacustris</i>	October 2001–July 2002	0.4	250	38	99	0.130	0.13	Søvik and Kløve (2007)
Domestic wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	August 2004–May 2006	0.0012	0.5	73.5	128	0.079	0.06	Gui et al. (2007)
Domestic wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	June–August 2006 33 days in summer	0.0012	0.5	100	174	0.2	0.12	Liu et al. (2009)
Agricul. non-point pollution	Mesocosms, USA	T	<i>P. australis</i> , <i>Typha</i> spp.	1998	n.a.	n.a.	n.a.	73	0.16	0.22	Xue et al. (1999)
Agricul. non-point pollution	Donaumoos, Germany	T	<i>Basin 1, T. latifolia</i> , <i>T. angustifolia</i>	September 1998–May 1999	2.2	158	51	15.3	0.009	0.06	Wild et al. (2001)
Agricul. non-point pollution	Donaumoos, Germany	T	<i>Basin 2, T. latifolia</i> , <i>T. angustifolia</i>	September 1998–May 1999	2.6	158	23	5.8	−0.003	−0.05	Wild et al. (2001)
Agricul. non-point pollution	Hovi, Finland	B	<i>T. latifolia</i> , <i>S. sylvaticus</i> , <i>A. plantago-aquatica</i> , <i>P. arundinacea</i> , <i>F. ulmaria</i> , <i>I. pseudacorus</i> , <i>J. conglomeratus</i>	January 2002–October 2003	0.6	78.6	1.4	1	0.001	0.07	Søvik et al. (2006)
Dairy farm wastewater	Truro Nova Scotia, Canada	T	<i>T. latifolia</i>	August 2005–September 2006	0.00066	0.105	306	203	0.25	0.12	Van der Zaag et al. (2010)
Raw municipal wastewater	Jiaonan, China	T	<i>P. australis</i> , <i>A. calamus</i> , <i>L. minor</i>	June 1999–August 2000	49.5	12,000	120	121	0.068	0.06	Tai et al. (2002)
Synthetic wastewater	Lab studies Jinan, China	W/T	<i>P. australis</i>	April–June 2008	0.000018	0.012	52	144	0.250	0.17	Wu et al. (2009)
Synthetic wastewater	Lab studies Jinan, China	W/T	<i>P. australis</i>	April–June 2008	0.000018	0.012	55	153	0.650 <sup>a</sup>	0.43 <sup>a</sup>	Wu et al. (2009)
Synthetic wastewater	Lab studies Jinan, China	W/T	<i>P. australis</i>	April–June 2008	0.000018	0.012	54	150	0.150	0.10	Wu et al. (2009)
Synthetic wastewater	Lab studies Jinan, China	W/T	<i>P. australis</i>	April–June 2008	0.000018	0.012	51	142	0.1	0.07	Wu et al. (2009)
Synthetic wastewater	Lab studies Jinan, China	W/T	<i>P. australis</i>	April–June 2008	0.000018	0.012	49	136	0.05	0.04	Wu et al. (2009)

<sup>a</sup> Not included in the correlation analysis (see text).

**Table 3**

Methane ( $\text{CH}_4\text{—C}$ ) and carbon dioxide ( $\text{CO}_2\text{—C}$ ) emission; inflow BOD or TOC values; inflow TOC loading ( $\text{TOC}_{\text{in}}$ ); and emission factor ( $\text{CH}_4\text{—C}/\text{TOC}_{\text{in}}$ ) in vertical subsurface (VSSF) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm. n.a. – not available.

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q(\text{m}^{-3} \text{d}^{-1})$	BOD ( $\text{mg L}^{-1}$ )	TOC ( $\text{mg L}^{-1}$ )	Inflow BOD load ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{TOC}_{\text{in}}$ ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CO}_2\text{—C}$ emiss. ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4\text{—C}$ flux ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4\text{—C}/\text{TOC}_{\text{in}}$ (%)	References
Domestic wastewater	Kõo, Estonia	T/B	<i>P. australis</i>	October 2001–November 2003	0.0128	65	32.2	16.1	681	341	208	3	0.88	Teiter and Mander (2005), Søvik et al. (2006) and Mander et al. (2008)
Domestic wastewater	Ski, Norway	B	No vegetation	June–December 2001	0.000005	0.042	n.a.	40.5	n.a.	1418	127	5.4	0.38	Søvik et al. (2006)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	August 2004–May 2006	0.0012	0.5	107.3	10.3	n.a.	17.9	n.a.	0.30	1.68	Gui et al. (2007)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	June–August 2006	0.0012	0.5	200	n.a.	347	174	n.a.	3	1.73	Liu et al. (2009)

**Table 4**

Nitrous oxide ( $\text{N}_2\text{O}\text{—N}$ ) emission; inflow total nitrogen loading ( $\text{TN}_{\text{in}}$ ); and emission factor ( $\text{N}_2\text{O}\text{—N}/\text{TN}_{\text{in}}$ ) in vertical subsurface (VSSF) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm.

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q(\text{m}^{-3} \text{d}^{-1})$	$\text{TN}(\text{mg L}^{-1})$	$\text{TN}_{\text{in}}$ ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{N}_2\text{O}\text{—N}$ flux ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{N}_2\text{O}\text{—N}/\text{TN}_{\text{in}}$ (%)	References
Domestic wastewater	Kõo, Estonia	T/B	<i>P. australis</i>	October 2001–November 2003	0.0128	65	50.9	1077	0.225	0.021	Teiter and Mander (2005) and Søvik et al. (2006)
Domestic wastewater	Ski, Norway	T/B	No vegetation	June–December 2001	0.000005	0.0042	52.6	1841	0.200	0.011	Søvik et al. (2006)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	August 2004–May 2006	0.0012	0.5	73.5	128	0.123	0.096	Gui et al. (2007)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	June–August 2006	0.0012	0.5	100	174	0.073	0.042	Liu et al. (2009)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	April 2001–December 2002	0.000025	0.17	9.8	278	0.003	0.001	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	April 2001–December 2002	0.000025	0.17	18.4	521	0.008	0.001	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	April 2001–December 2002	0.000025	0.17	36.7	1040	0.033	0.003	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>Zizania latifolia</i>	April 2001–December 2002	0.000025	0.17	9.8	278	0.005	0.002	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>Z. latifolia</i>	April 2001–December 2002	0.000025	0.17	18.4	521	0.023	0.004	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>Z. latifolia</i>	April 2001–December 2002	0.000025	0.17	36.7	1040	0.040	0.004	Inamori et al. (2007)
Artificial wastewater	Ibaraki, Japan	T/W	<i>T. latifolia</i>	January 2005–September 2006	0.000025	0.421	15	1053	0.200	0.019	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>T. latifolia</i>	January 2005–September 2006	0.000025	0.421	30	2105	0.353	0.017	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>T. latifolia</i>	January 2005–September 2006	0.000025	0.421	15	1053	0.211	0.020	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>T. latifolia</i>	January 2005–September 2006	0.000025	0.421	30	2105	0.424	0.020	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	January 2005–September 2006	0.000025	0.421	15	1053	0.188	0.018	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i>	January 2005–September 2006	0.000025	0.421	30	2105	0.371	0.018	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	Non-vegetated control	January 2005–September 2006	0.000025	0.421	15	1053	0.150	0.014	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	Non-vegetated control	January 2005–September 2006	0.000025	0.421	30	2105	0.162	0.008	Inamori et al. (2008)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i> , <i>T. latifolia</i> , <i>Z. latifolia</i>	January 2005–August 2006	0.000025	0.17	3.62	103	0.034	0.033	Wang et al. (2008a,b)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i> , <i>T. latifolia</i>	January 2005–August 2006	0.000025	0.17	6.65	188	0.110	0.058	Wang et al. (2008a,b)
Artificial wastewater	Ibaraki, Japan	T/W	<i>P. australis</i> , <i>Z. latifolia</i>	January 2005–August 2006	0.000025	0.17	4.5	128	0.043	0.034	Wang et al. (2008a,b)
Artificial wastewater	Ibaraki, Japan	T/W	Non-vegetated control	January 2005–August 2006	0.000025	0.17	10	283	0.073	0.026	Wang et al. (2008a,b)

**Table 5**

Methane ( $\text{CH}_4$ —C) and carbon dioxide ( $\text{CO}_2$ —C) emission; inflow BOD or TOC values; inflow TOC loading ( $\text{TOC}_{\text{in}}$ ); and emission factor ( $\text{CH}_4$ —C/ $\text{TOC}_{\text{in}}$ ) in horizontal subsurface (HSSF) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm, M – Mediterranean, Tr – tropical. n.a. – not available.

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	$Q(\text{m}^{-3} \text{d}^{-1})$	BOD ( $\text{mg L}^{-1}$ )	TOC ( $\text{mg L}^{-1}$ )	Inflow BOD load ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{TOC}_{\text{in}} (\text{mg m}^{-2} \text{h}^{-1})$	$\text{CO}_2$ —C emission ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4$ —C flux ( $\text{mg m}^{-2} \text{h}^{-1}$ )	$\text{CH}_4$ —C/ $\text{TOC}_{\text{in}}$ (%)	References
Domestic wastewater	Kodijärve, Estonia	T/B	<i>T. latifolia</i> , <i>P. australis</i> , <i>Scirpus sylvaticus</i>	October 2001–November 2003	0.03125	2.85	n.a.	69	n.a.	26	99.2	2.6	9.9	Mander et al. (2003, 2005a,b, 2008), Teiter and Mander (2005), Søvik et al. (2006)
Domestic wastewater	Kõo, Estonia	T/B	<i>P. australis</i>	October 2001–November 2003	0.0365	60	62.8	n.a.	430	215	41.7	3.09	1.4	Teiter and Mander (2005) and Søvik et al. (2006)
Domestic wastewater	Ski, Norway	B	No vegetation	June–December 2001	0.00009	0.072	n.a.	22.3	n.a.	74	51.9	7.1	9.6	Søvik et al. (2006)
Domestic wastewater	Barcelona, Spain	T/M	Non-vegetated gravel from a CW	20 days lab exper. in gravel beds	0.0055	1.98	200	n.a.	300	150	128	0.048	0.03	Garcia et al. (2007)
Domestic wastewater	Miho, Ibaraki Japan	T/W	<i>P. australis</i>	August 2004–May 2006	0.0012	0.5	107.3	10.3	n.a.	17.9	n.a.,	0.76	4.3	Gui et al. (2007)
Domestic wastewater	Slavosovice Czech Republic	T	<i>P. australis</i>	June–October 2004	0.0748	10.4	n.a.	38.1	n.a.	22.1	174.7	17.5 <sup>a</sup>	79.3 <sup>a</sup>	Picek et al. (2007)
Domestic wastewater	Miho, Ibaraki, Japan	T/W	<i>P. australis</i>	June–August 2006	0.0012	0.5	200	n.a.	347	174	n.a.	7	4	Liu et al. (2009)
Dairy farm wastewater	Truro, Nova Scotia, Canada	T	<i>T. latifolia</i>	August 2005–September 2006	0.00066	0.105	186	n.a.	123.3	61.6	146	4.9	7.9	Van der Zaag et al. (2010)
Peat mining runoff	Kompsasuo Finland	B	<i>Sphagnum angustifolium</i> , <i>S. papillosum</i> , <i>M. trifoliata</i> , <i>Carex spp.</i>	June–August 1992	1.9	n.a.	n.a.	n.a.	n.a.	16.9	303	5.8	34.3 <sup>a</sup>	Liikanen et al. (2006)
Peat mining runoff	Kompsasuo Finland	B	<i>M. trifoliata</i> , <i>C. lasiocarpa</i> , <i>P. palustris</i> , <i>Sphagn spp.</i>	August 2001–August 2002	1.9	n.a.	n.a.	n.a.	n.a.	15	567	16.7	111.3 <sup>a</sup>	Liikanen et al. (2006)
Peat mining runoff	Kompsasuo, Finland	B	<i>Sphagn spp.</i> , <i>Menyanthes trifoliata</i> , <i>Carex lasiocarpa</i> , <i>Potentilla palustris</i>	January 2002–October 2003	2.4	1857	n.a.	15.0	n.a.	8.2	n.a.	13.1 <sup>a</sup>	160 <sup>a</sup>	Søvik et al. (2006)
Landfill leachate fresh	Bangkok, Thailand	Tr	<i>T. angustifolia</i>	January–December 2006	0.0003	0.056	5275	2816		2190 <sup>a</sup>	n.a.	14.3	0.7	Chiemchaisri et al. (2008)
Landfill leachate stabilized	Bangkok, Thailand	TrI	<i>T. angustifolia</i>	January–December 2006	0.0003	0.056	42	403		313	n.a.	9.25	3.0	Chiemchaisri et al. (2008)

<sup>a</sup> Not included in the analysis (see text).

farm wastewater treatment in Friedelhausen, Germany (Fey et al., 1999) has also been included (Table 6).

Tanner et al. (1997) presented estimated values for inflow of total organic carbon (TOC<sub>in</sub>), Xue et al. (1999) for inflow total nitrogen (TN<sub>in</sub>), and Sovik et al. (2006) for both TOC<sub>in</sub> and TN<sub>in</sub>. For most of the systems, TOC<sub>in</sub> and TN<sub>in</sub> values in g m<sup>-2</sup> h<sup>-1</sup> were calculated based on area, hydraulic load and inflow TOC or BOD and TN concentration data.

The EF values were calculated as:

$$EF_{CH_4} = (CH_4 - C_{\text{emission}} / TOC_{in}) * 100(\%) \quad (1)$$

$$EF_{N_2O} = (N_2O - N_{\text{emission}} / TN_{in}) * 100(\%) \quad (2)$$

The TOC<sub>in</sub> values were available for 8 FWS CWs (Table 1), for all four VSSF CWs (Table 3), and for 6 HSSF CWs (Table 5). For the rest of the systems only biochemical oxygen demand (BOD) values were usable, and for these the following approximation based on domestic wastewater data was used: TOC = 0.5 BOD (Garcia et al., 2007).

For the calculation of emission factors, we used data series from one year or at least a vegetation period. Some extreme values of CH<sub>4</sub> emissions and related EF values from Jiaonan, Skjønhaug (Table 1), Kompsasuo and Slavosovice (Table 5), as well as one extreme value for N<sub>2</sub>O emission by Wu et al. (2009) (Table 2) were not used in further analyses. Likewise, we did not include in our study CH<sub>4</sub> emission values from small-scale laboratory experiments by Inamori et al. (2007), Wang et al. (2008a,b), and Maltais-Landry et al. (2009a,b).

All of the GHG emission data in the analyzed publications were collected using the static chamber/gas-chromatograph technique (Hutchinson and Livingston, 1993). No publications could be found on the use of eddy covariance technique for measuring GHG fluxes in CWs.

## 2.2. Statistical analysis of data

The STATISTICA 7.1 program was used for the data analysis. We tested the normality of variables with Lilliefors' and Shapiro-Wilk's tests. All variables were log-transformed prior to data analysis. Differences in average CH<sub>4</sub> and N<sub>2</sub>O emission values between wetland types and due to the presence of vegetation were tested using one-way analysis of variance (ANOVA). Stepwise multiple regression analysis with the forward selection option was applied to select predictor variables of CH<sub>4</sub> and N<sub>2</sub>O emissions in the case of a particular wetland type. The level of significance  $\alpha = 0.05$  was accepted in all cases.

## 3. Results

### 3.1. Carbon dioxide emission

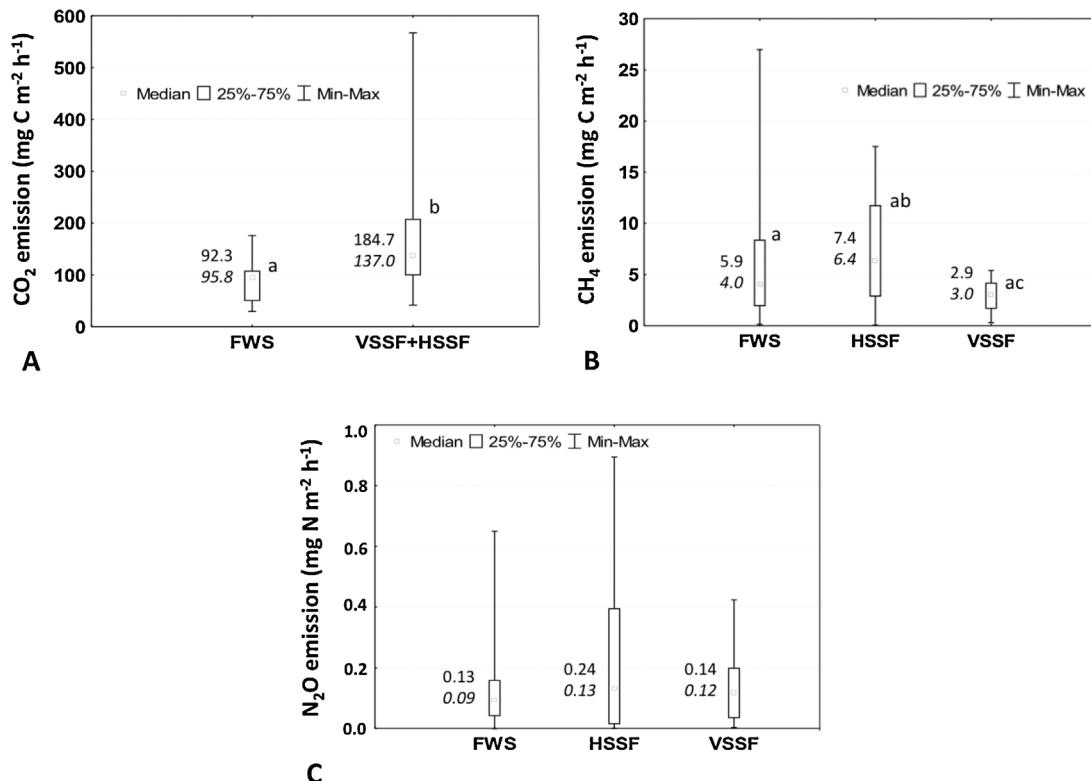
The emission of CO<sub>2</sub>-C was significantly lower in FWS CWs (ranging from 29.4 to 176.0, with average and median values of 92.3 and 95.8 mg m<sup>-2</sup> h<sup>-1</sup>, respectively) than in subsurface flow CWs (VSSF + HSSF; 51.9–567.0; average 184.7, median 137 mg m<sup>-2</sup> h<sup>-1</sup>; Fig. 1A; Tables 1, 3 and 5).

There is a significant negative correlation between the inflow TOC loading (TOC<sub>in</sub>) value and CO<sub>2</sub>-C emission (Fig. 2A).

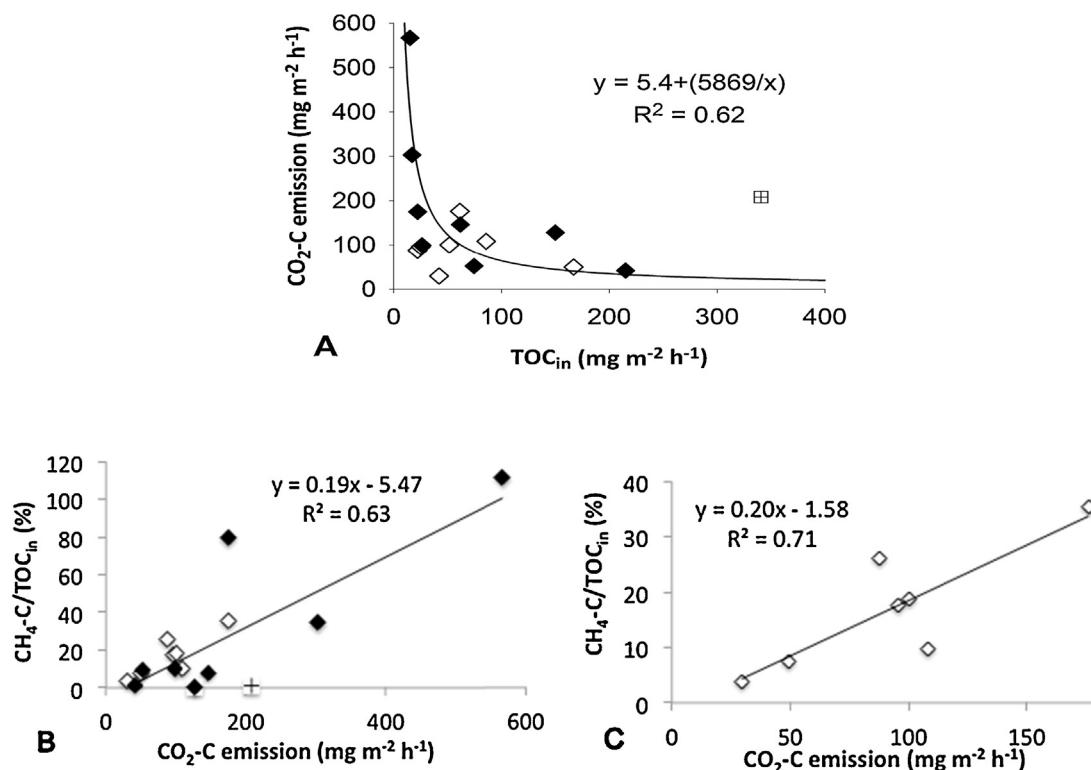
Likewise, we have found a positive correlation ( $p < 0.05$ ) between the CO<sub>2</sub>-C emission and CH<sub>4</sub> emission factor (CH<sub>4</sub>-C/TOC<sub>in</sub>) in all CW types (Fig. 2B), whereas this correlation was stronger ( $p < 0.01$ ) in FWS CWs (Fig. 2C).

**Table 6**  
Nitrous oxide (N<sub>2</sub>O-N) emission; inflow total nitrogen loading (TN<sub>in</sub>); and emission factor (N<sub>2</sub>O-N/TN<sub>in</sub>) in horizontal subsurface (HSSF) constructed wetlands for wastewater treatment. Average values for each site/event are presented. Climate zones: T – temperate, B – boreal, W – warm, n.a. – not available.

Waste-water type	Study site, country	Climate zone	Plant species	Period of measurement	Area (ha)	Q/(m <sup>-3</sup> d <sup>-1</sup> )	TN (mg L <sup>-1</sup> )	TN <sub>in</sub> (mg m <sup>-2</sup> h <sup>-1</sup> )	N <sub>2</sub> O–N/flux (mg m <sup>-2</sup> h <sup>-1</sup> )	N <sub>2</sub> O–N/TN <sub>in</sub> (%)	References
Domestic wastewater	Kodijärvi, Estonia	T/B	<i>Typha latifolia</i> , <i>Phragmites australis</i> , <i>Schoenus sylvaticus</i> <i>P. australis</i>	October 2001–November 2003	0.03125	2.85	109	41	0.186	0.45	Mander et al. (2003, 2005a,b, 2008) and Teiter and Mander (2005)
Domestic wastewater	KÖÖ, Estonia	T/B		October 2001–November 2003	0.0365	60	43.1	295	0.108	0.04	Teiter and Mander (2005), Sovik et al. (2006) and Mander et al. (2008)
Domestic wastewater	Ski, Norway	B	Non-vegetated	June–December 2001	0.00009	0.012	53.4	30	0.894	3.01	Sovik et al. (2006)
Domestic wastewater	Miho Ibaraki Japan	T/W	<i>P. australis</i> <i>Phragmites australis</i>	June–August 2006	0.0012	0.5	100	174	0.4	0.23	Liu et al. (2009)
Dairy farm wastewater	Friedelhausen, Germany	T		November 1995–March 1996	0.018	12	73	203	0.133	0.07	Fey et al. (1998)
Dairy farm wastewater	Truro, Nova Scotia, Canada	T	<i>T. latifolia</i>	August 2005–September 2006	0.00066	0.105	306	202.8	0.396	0.20	Van der Zaag et al. (2010)
Peat mining runoff	Kompsasuo, Finland	B	<i>Sphagnum</i> spp., <i>M. trifoliate</i> , <i>C. fastigiatum</i> , <i>P. palustris</i> <i>S. angustifolium</i> , <i>S. papillosum</i> , <i>M. trifoliate</i> , <i>Carex</i> spp. <i>M. trifoliate</i> , <i>C. lasiocarpa</i> , <i>P. palustris</i> , <i>Sphagnum</i> spp.	January 2002–October 2003	0.6	78.6	1.8	1	0.005	0.47	Sovik et al. (2006)
Peat mining runoff	Kompsasuo, Finland	B		June–August 1992	1.9	n.a.	n.a.	3.04	0.014	0.46	Liikonen et al. (2006)
Peat mining runoff	Kompsasuo, Finland	B		August 2001–August 2002	1.9	n.a.	n.a.	1.04	0.019	1.83	Liikonen et al. (2006)



**Fig. 1.** Average, median, 25% and 75% quartile and min/max values of carbon dioxide (A), methane (B), and nitrous oxide (C) emissions in various types of constructed wetlands (CW). FWS – free water surface, VSSF – vertical subsurface flow, HSSF – horizontal subsurface flow CWs. Numbers close to boxes indicate average values (above) and median values (below, in italic). Different lower case letters indicate significant differences in emission values between various types of CWs ( $p < 0.05$ ).



**Fig. 2.** The relationship between inflow TOC loading (TOC<sub>in</sub>) and CO<sub>2</sub>-C emission in all types of CWs studied (A), and between the CO<sub>2</sub>-C emission and CH<sub>4</sub> emission factor (CH<sub>4</sub>-C/TOC<sub>in</sub>) for all types of CWs (B) and in FWS CWs (C). White diamonds – FWS, black diamonds – HSSF, and crosses – VSSF CWs. In A and B:  $p < 0.05$ , in C:  $p < 0.01$ .

**Table 7**

Relationships between CH<sub>4</sub> and N<sub>2</sub>O emission and wetland operational parameters (TOC<sub>in</sub>, TN<sub>in</sub>, Q) based on stepwise multiple regression analysis.

Dependent variable	Wetland type	Variables in model	Regression coefficient	R <sup>2</sup> value (%)	p Value
CH <sub>4</sub> emission	FWS	TOC <sub>in</sub>	0.58	30.0	<0.001
CH <sub>4</sub> emission	HSSF	TOC <sub>in</sub>	0.34	40.4	<0.05
CH <sub>4</sub> emission	HSSF	Q	-0.31	40.4	<0.01
N <sub>2</sub> O emission	FWS	TN <sub>in</sub>	0.02	23.7	<0.05
N <sub>2</sub> O emission	VSSF	TN <sub>in</sub>	0.07	43.9	<0.001
N <sub>2</sub> O emission	HSSF	TN <sub>in</sub>	0.06	23.9	<0.05

### 3.2. Methane emission

In VSSF CWs the CH<sub>4</sub>-C emission (range 0.3–5.4, average 2.9, median 3.0 mg m<sup>-2</sup> h<sup>-1</sup>) was significantly lower than in HSSF CWs (0.048–17.5, 7.4 and 6.4 mg m<sup>-2</sup> h<sup>-1</sup>) and FWS CWs (0.15–27, 5.9 and 4.0 mg m<sup>-2</sup> h<sup>-1</sup>) (Fig. 1B). Multiple regression analysis yielded a significant correlation between the TOC<sub>in</sub> and CH<sub>4</sub>-C emission values in all types of CWs (Table 7 and Fig. 3).

Significant differences have been found between the EF values in different CW types. The highest value was found for FWS CWs, in which on average 16.9% of inflow TOC is transformed into CH<sub>4</sub>-C (median value being 18.0; Table 8). This is followed by HSSF CWs, where the average and median EF values are 4.5% and 3.8%, respectively, whereas in VSSF CWs only 1.17% (median value 1.28%) of TOC<sub>in</sub> is transformed to methane (Table 8).

### 3.3. Nitrous oxide emission

There were no significant differences in N<sub>2</sub>O-N emissions in various CW types (Table 8). The lowest values were found in FWS CWs (ranging from -0.003 to 0.65; average 0.13, median 0.09 mg m<sup>-2</sup> h<sup>-1</sup>), followed by VSSF CWs (0.003–0.424; average 0.14, median 0.12 mg m<sup>-2</sup> h<sup>-1</sup>) and HSSF CWs (0–0.894, average 0.24, median 0.13 mg m<sup>-2</sup> h<sup>-1</sup>) (Fig. 1C). All of the CW types showed a significant positive correlation between the inflow TN loading and N<sub>2</sub>O-N emission values (Table 8 and Fig. 4).

Average N<sub>2</sub>O emission factors differed significantly in all three CW types: 0.79% (median 0.34%) in HSSF, 0.13% (median 0.11%) in FWS, and 0.023% (median 0.018%) in VSSF CWs (Table 9).

## 4. Discussion

### 4.1. Carbon dioxide emission

Although CO<sub>2</sub> is a major GHG, there are only a limited number of studies reporting on CO<sub>2</sub> emissions in CWs (Mander et al., 2003a,b, 2005a,b, 2008; Teiter and Mander, 2005; Liikanen et al., 2006; Garcia et al., 2007; Søvik et al., 2006; Van der Zaag et al., 2010). The values of CO<sub>2</sub> emissions (soil + root + rhizome respiration + litter decomposition) vary from 0.88 to 2.37 kg CO<sub>2</sub>-Cha<sup>-1</sup> yr<sup>-1</sup> (Garcia et al., 2007) in anaerobic HSSF CWs, up to 3840–7360 kg CO<sub>2</sub>-Cha<sup>-1</sup> yr<sup>-1</sup> in a well-aerated HSSF CW with a lowered water table and well-developed macrophyte cover (*Scirpus sylvaticus*, *Phragmites australis*) (Mander et al., 2008). Some of the emitted CO<sub>2</sub> will be assimilated by plants, and in optimal conditions the carbon sequestration in the HSSF CW filter material can reach 15,500–20,750 kg CO<sub>2</sub>-Cha<sup>-1</sup> yr<sup>-1</sup> (Mander et al., 2008). This carbon is, however, non-stable and can be easily mineralized when the water table lowers or when the filter material, which becomes saturated with phosphorus, is replaced with a new one.

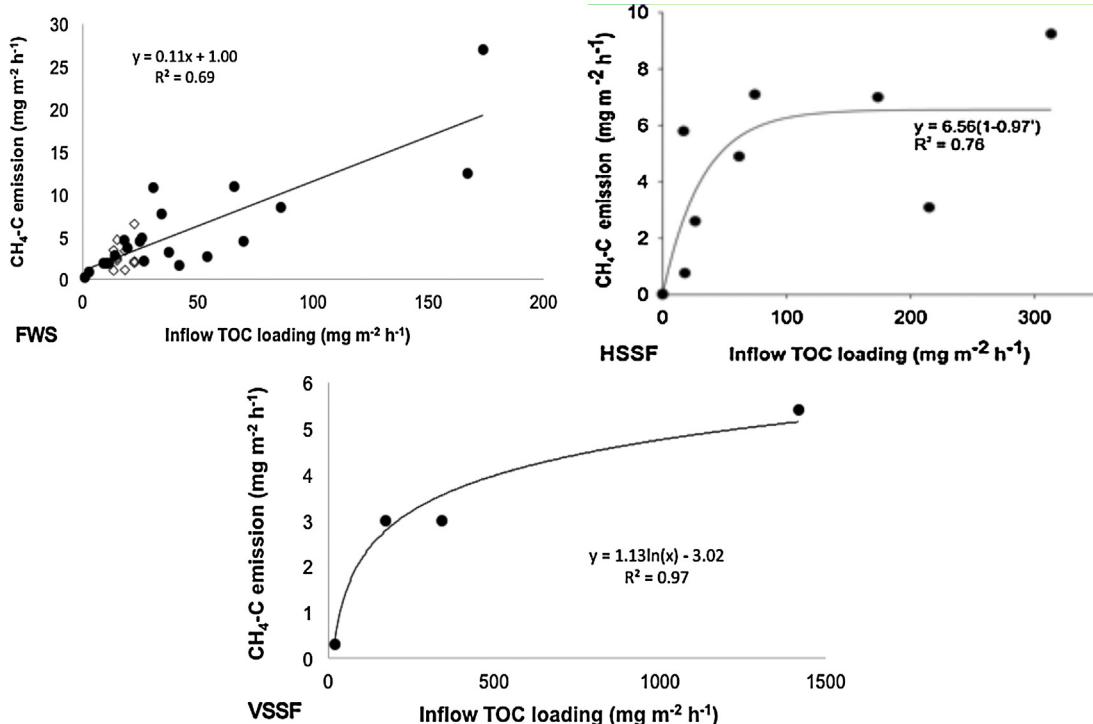


Fig. 3. The relationship between inflow TOC loading (TOC<sub>in</sub>) and CH<sub>4</sub>-C emission in FWS, VSSF and HSSF CWs (p < 0.05).

**Table 8**

Emission factors of methane and nitrous oxide in treatment wetlands: share (%) of CH<sub>4</sub>—C emission in the inflow loading of TOC (TOC<sub>in</sub>) and N<sub>2</sub>O—N emission in the inflow loading of TN (TN<sub>in</sub>). The letters a–c and x–z indicate significant ( $p < 0.05$ ) differences between CH<sub>4</sub> and N<sub>2</sub>O emission factors, respectively.

Type of wetland	CH <sub>4</sub> —C/TOC <sub>in</sub> (%)				N <sub>2</sub> O—N/TN <sub>in</sub> (%)			
	Average	Median	Standard Error	Number of analyses	Average	Median	Standard Error	Number of analyses
Free water surface (FWS) wetlands	16.9 <sup>a</sup>	18.0	1.8	22	0.13 <sup>x</sup>	0.11	0.024	24
Horizontal subsurface flow (HSSF) wetlands	4.5 <sup>b</sup>	3.8	1.1	9	0.79 <sup>y</sup>	0.34	0.38	8
Vertical subsurface flow (VSSF) wetlands	1.17 <sup>c</sup>	1.28	0.33	4	0.023 <sup>z</sup>	0.018	0.005	22

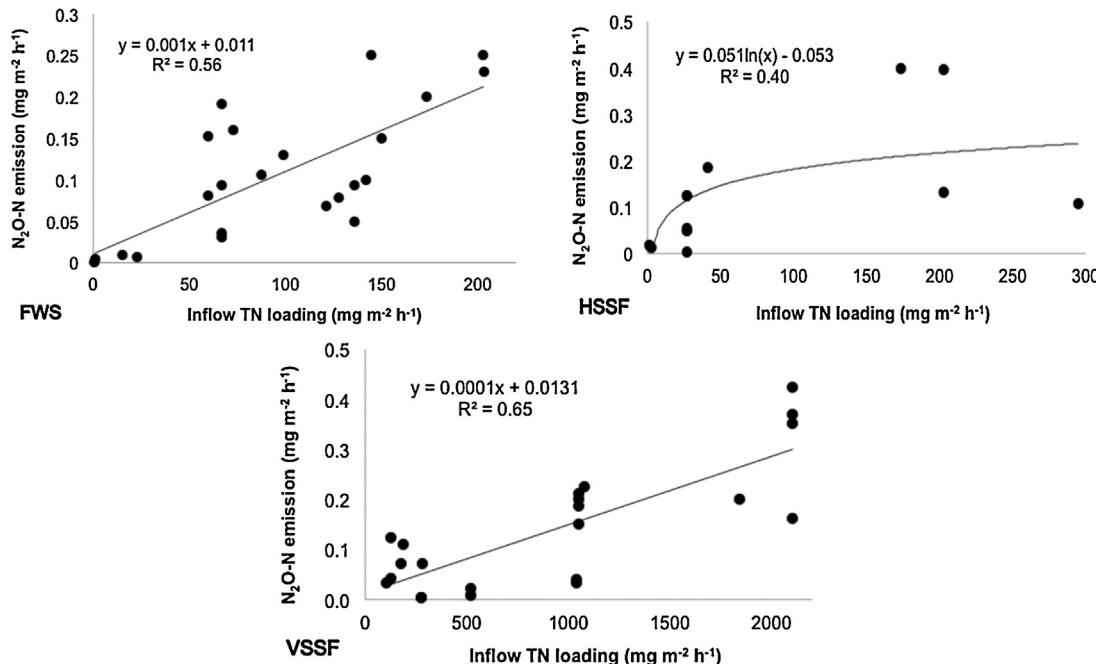


Fig. 4. The relationship between inflow TN loading (TN<sub>in</sub>) and N<sub>2</sub>O—N emission in FWS, VSSF, and HSSF CWs ( $p < 0.05$ ).

**Table 9**

Selected factors and processes influencing CH<sub>4</sub>, and N<sub>2</sub>O emission in constructed wetlands.

Influencing factors/processes	CH <sub>4</sub> emission	N <sub>2</sub> O emission
Higher water/soil/air temperature	Increase in almost all cases <sup>a,b,c,d,e,f</sup> with few exceptions <sup>g</sup>	No clear relationship <sup>a,b,c,d,g,h</sup>
Higher moisture in soil or filter material (higher value of WFPS)	Clear increase <sup>i,j</sup>	Decrease <sup>i,j</sup>
Higher wastewater loading	Increase <sup>a,b,c,d,k,l,m</sup>	Increase <sup>a,b,d,n</sup>
Presence of aerenchymal plants	Increase <sup>o,p,q</sup> /decrease (depends on conditions) <sup>f</sup>	Increase <sup>s</sup> /decrease <sup>q,t</sup>
Pulsing hydrological regime (intermittent loading)	Clear decrease <sup>i,u</sup>	Increase <sup>i,v,w</sup> /decrease in some SF CWs <sup>x</sup>
Deeper water table (from surface) in HSSF CWs	Decrease <sup>i,j</sup>	Increase <sup>i,j</sup>

<sup>a</sup> Mander et al. (2003).

<sup>b</sup> Mander et al. (2005a,b).

<sup>c</sup> Teiter and Mander (2005).

<sup>d</sup> Søvik et al. (2006).

<sup>e</sup> Kayranli et al. (2010).

<sup>f</sup> Van der Zaag et al. (2010).

<sup>g</sup> Søvik and Kløve (2007).

<sup>h</sup> Fey et al. (1999).

<sup>i</sup> Mander et al. (2011).

<sup>j</sup> Yang et al. (2013).

<sup>k</sup> Tanner et al. (1997).

<sup>l</sup> Tai et al. (2002).

<sup>m</sup> Picek et al. (2007).

<sup>n</sup> Hunt et al. (2009).

<sup>o</sup> Inamori et al. (2007).

<sup>p</sup> Inamori et al. (2008).

<sup>q</sup> Wang et al. (2008a,b).

<sup>r</sup> Maltais-Landry et al. (2009a,b).

<sup>s</sup> Rückauf et al. (2004).

<sup>t</sup> Silvan et al. (2005).

<sup>u</sup> Altor and Mitsch (2006).

<sup>v</sup> Jia et al. (2011).

<sup>w</sup> Van de Riet et al. (2013).

<sup>x</sup> Hernandez and Mitsch (2006).

Our analysis showed that CO<sub>2</sub> emission from CWs is negatively correlated with TOC<sub>in</sub> in HSSF CWs (Fig. 2A). Above a certain TOC<sub>in</sub> level ( $>100 \text{ mg C m}^{-2} \text{ h}^{-1}$ ) the treatment wetlands do not more than 200 mg CO<sub>2</sub>–C m<sup>-2</sup> h<sup>-1</sup>, which indicate that above that TOC<sub>in</sub> value the purification process also no longer intensifies. Likewise, CH<sub>4</sub> emission is stabilized above the TOC<sub>in</sub> value  $>100 \text{ mg C m}^{-2} \text{ h}^{-1}$  (Fig. 3).

The CO<sub>2</sub> emission factor (CO<sub>2</sub>–C/TOC<sub>in</sub>) varied from 9% to 3780%, and was in most cases higher than 100%. The smallest CO<sub>2</sub> EF values were measured in VSSFs and HSSFs with scarce plant cover (Tables 1, 3 and 5). This clearly shows the significant role of aquatic plants, which bring a great deal of atmospheric CO<sub>2</sub> into the system, thereby establishing the basis for C sequestration (Mitsch and Gosselink, 2007).

Eddy covariance data were not available in CWs, although the studies conducted in an abandoned agricultural peat meadow, which can be considered to be a proxy ecosystem to HSSF CWs, showed a significant variation in net ecosystem exchange (NEE), namely  $-446$  and  $-232 \text{ g C m}^{-2} \text{ yr}^{-1}$  in wet and dry years, respectively. CO<sub>2</sub> emission has decreased significantly as result of the raised water table, while CH<sub>4</sub> fluxes have increased. Over the whole study period (2004–2006), the area was a small net GHG sink, given as CO<sub>2</sub>-eq. of  $-86 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Hendriks et al., 2007).

Nevertheless, with the right design takes into consideration all of the regulation and supporting services of CW systems, it would be possible to achieve significant C sequestration such as that which takes place in natural wetlands (Mitsch and Gosselink, 2007; Mitsch et al., 2013).

## 4.2. Methane emission

Methane seems to be the most important GHG emitted from CWs. In FWS CWs an average of 16.9% of TOC<sub>in</sub> is transformed to CH<sub>4</sub>–C (Table 8). This is due to the intensive accumulation of organic matter (OM) in CWs (Mander et al., 2014). Some of the formed methane is probably oxidised and emitted as CO<sub>2</sub>, which is supported by a significant correlation of CO<sub>2</sub>–C emission and a CH<sub>4</sub> EF value (CH<sub>4</sub>–C/TOC; Fig. 2B and C). The non-linear character of CH<sub>4</sub>–C emission vs. the TOC<sub>in</sub> relationship in HSSF and VSSF CWs (Fig. 3) may indicate some limit level for CH<sub>4</sub> release, i.e., from a certain TOC<sub>in</sub> loading, methanogenesis may be inhibited by increased concentration of ammonia, or accumulation of volatile fatty acids inhibiting the rate limiting step of methanogenesis, hydrolysis of organic matter (Mata-Alvarez et al., 2000). However, the limited amount of data available prevented us from drawing more comprehensive conclusions. On the other hand, excessively heavy loading leads to the clogging of the system, requiring corresponding management activity. Also, very high overloading, such as in the case of FWS CWs, rarely happens in practice.

Overloaded FWS CWs such as the Jiaonan wetland system for raw wastewater treatment (Tai et al., 2002; Table 1) and the Skjønhaug wetland (Søvik and Kløve, 2007; Table 1) with a high OM accumulation rate had EF values that even exceeded 100%. Likewise, the Slavosovice HSSF CW (Picek et al., 2007; Table 5) showed very high EF values (79%), which is probably due to overloading and clogging problems in the system. The Kompasuo peat mining wastewater treatment system in a Finnish peatland is a mixture of HSSF and FWS CWs (Liikanen et al., 2006), and is therefore considered under both categories (Søvik et al., 2006) or as an FWS CW (Mander et al., 2013). In this paper we follow the approach proposed by Liikanen et al. (2006) and consider the Kompasuo treatment system as an HSSF CW (Table 5). This system does, however, also generate high values of CH<sub>4</sub> emission, whereas during the period from 1992 to 2002, EF increased from 34.3% to 111.3% (Liikanen et al., 2006; Table 5). For the period 2002–2003,

calculations based on the data of Søvik et al. (2006) give an EF value as high as 160% (Table 5). The data from all of the over-loaded systems described above, as well as from the peat extraction wastewater treatment systems, were not included in our further analysis.

## 4.3. Nitrous oxide emission

Findings from earlier studies on GHG emissions in HSSF and VSSF CWs which show high N<sub>2</sub> and low N<sub>2</sub>O emission – indicators of almost complete denitrification – suggest denitrification might be the main source of nitrous oxide (Mander et al., 2003, 2005a, 2008; Teiter and Mander, 2005). Heterotrophic denitrification relies on the absence of O<sub>2</sub> for the conversion of N<sub>2</sub>O to N<sub>2</sub>; if oxygen is introduced during this last step, incomplete denitrification will occur and N<sub>2</sub>O will be released as the end product instead of N<sub>2</sub>. In wetland systems, where both ammonia and nitrate are present, the intermittent loading in VSSF CWs (Jia et al., 2011) and the fluctuating water table in HSSF CWs (Mander et al., 2011) which bring intermittent oxygen to the system can result in increased emissions by affecting both nitrification (through nitrifier stress, thus triggering nitrifier denitrification) and denitrification (by interrupting the last biochemical step; Dotro et al., 2011). The introduction of artificial aeration in laboratory-scale HSSF CWs (Maltais-Landry et al., 2009a,b) resulted in increased emissions; it is, however, unclear if this was due to greater air stripping of the dissolved gas or as a result of increased nitrification rates. In a laboratory-scale VSSF CW, Zhou et al. (2008) found a significant positive correlation between the N<sub>2</sub>O–N flux and redox potential, suggesting nitrification was the dominant N<sub>2</sub>O-emitting microbial process. Yu et al. (2006) found that redox potential was a significant indicator of N<sub>2</sub>O emission in soils of Gulf coast forests: maximum N<sub>2</sub>O concentration in these soils was found at about Eh +250 mV. However, additional measurement of N<sub>2</sub> emissions and isotope analysis are needed to confirm these statements.

The character of N<sub>2</sub>O–N emission vs. the TN<sub>in</sub> relationship (Fig. 4) may also support the denitrification hypothesis: in contrast to the linear correlation in FWS and VSSF CWs, the relation is non-linear in HSSF CWs, in which dominating anaerobic conditions support complete denitrification. Higher than  $50 \text{ mg N m}^{-2} \text{ hr}^{-1}$  of TN<sub>in</sub> loading (Fig. 4) may enhance the anaerobic conditions in HSSFs, and if there is enough carbon in the system (which is a common case in HSSFs) the denitrification might be completed and the main product will be N<sub>2</sub>.

Likewise, studies on the filling modes of a sequencing batch reactor (SBR) demonstrated that a major emission of N<sub>2</sub>O took place at the aerobic phase, while N<sub>2</sub>O emission at the anoxic phase was insignificant (Park et al., 2001).

## 4.4. Impact of physical, hydrological and operational factors

Table 9 presents a brief overview of selected factors and processes that control CH<sub>4</sub> and N<sub>2</sub>O emission in CWs. Higher inflow loading of both TOC and TN always increases the respective CH<sub>4</sub> and N<sub>2</sub>O fluxes. This is also demonstrated by the significant correlation between the inflow TOC and TN values and the corresponding CH<sub>4</sub> and N<sub>2</sub>O fluxes (Figs. 3 and 4). In most cases, the higher value of water-filled pore space (WFPS) of soils/sediments or the higher moisture of the filter material increased CH<sub>4</sub> emission and decreased N<sub>2</sub>O emission. Likewise, the deeper water table (from surface) in HSSF CWs always decreased CH<sub>4</sub> emission and increased N<sub>2</sub>O emission (Table 9). In arable soils, however, the highest N<sub>2</sub>O emission was found at WFPS values between 50% and 80% (Vilain et al., 2010). With few exceptions (Søvik and Kløve, 2007), the higher temperature of the environment always increased CH<sub>4</sub>

emission, whereas in terms of N<sub>2</sub>O the relationship is unclear. Our analysis along all CW types did not yield a significant relationship with CH<sub>4</sub> and N<sub>2</sub>O emission and climate zone (see Tables 1–6), although there was a slight increasing trend in CH<sub>4</sub> emission toward boreal CWs.

Wu et al. (2009) demonstrate that in FWS CWs the higher inflow COD concentration causes a significant increase in N<sub>2</sub>O emission, although our review analysis was unable to find a significant correlation between TOC<sub>in</sub> and N<sub>2</sub>O emission.

The location of sampling sites in the flow path is an additional important factor for GHG emission. In most cases, the CH<sub>4</sub> emission in the inflow part of FWS and HSSF CWs has been significantly higher than in the outflow part (Tanner et al., 1997; Mander et al., 2005a,b; Søvik et al., 2006; Picek et al., 2007); some investigations have noted the same effect regarding the N<sub>2</sub>O flux (Mander et al., 2003) for an HSSF CW; Hernandez and Mitsch (2006) and Pulou (2011) for artificial riverine wetlands), and some for both CH<sub>4</sub> and N<sub>2</sub>O emission (Mander et al., 2005a,b; Teiter and Mander, 2005; Søvik et al., 2006).

The ratio of soil/sediment C/N is an important ecosystem parameter controlling many processes (Kalbitz et al., 2000). Yan et al. (2012) determined that the optimum C/N ratio is 5:1, at which point VSSF CWs can achieve relatively high biological nutrient removal efficiency and a low level of CO<sub>2</sub> and CH<sub>4</sub> emission. A low C/N ratio may, however, significantly increase N<sub>2</sub>O emission. This has been shown to occur in forested histosols (Klemmedsson et al., 2005) and activated sludge plants for domestic wastewater treatment, where 20–30% of the influent N was converted to N<sub>2</sub>O at a C/N ratio below 3.5 (Itokawa et al., 2001). Van der Zaag et al. (2010) report the same phenomenon for FWS and HSSF CWs: N<sub>2</sub>O emissions increased when influent wastewater had a low C/N ratio. This shift toward increased N<sub>2</sub>O emission at lower C/N ratio values may be due to alterations in the structure of the microbial community involved in nitrogen transformation in CW, and particularly in denitrifying microbial species. A low C/N ratio may favor microbial species with incomplete denitrification pathways (lack of nitrous oxide reductase encoding nosZ gene), which leads to an increase in N<sub>2</sub>O as an end product of denitrification.

It is known that iron and sulphur compounds significantly suppress CH<sub>4</sub> emission in peatlands (Dise et al., 1993). Decreases in CH<sub>4</sub> emissions measured after the addition of Fe<sup>3+</sup> to paddy soils (Jäckel and Schnell, 2000; Huang et al., 2009) and SO<sub>2</sub><sup>-4</sup> to rice field soils (Van der Gon and Neue, 1994) and histosols (Dise and Verry, 2001; Gauci et al., 2002) have been attributed to the inhibition of methanogenesis, predominantly through the stimulation of SO<sub>2</sub><sup>-4</sup> and Fe<sup>3+</sup> reducing bacteria which out-compete methanogens for acetate and hydrogen. Stimulation of CH<sub>4</sub> oxidation by SO<sub>2</sub><sup>-4</sup> (under anaerobic and aerobic conditions) and Fe<sup>3+</sup> addition (under anaerobic conditions) may also contribute to these observed decreases in CH<sub>4</sub> emissions (Kumaraswamy et al., 2001). On the other hand, in laboratory conditions Fe<sup>3+</sup> application to paddy soil increased N<sub>2</sub>O emission (Huang et al., 2009).

However, there are only few examples on the effect of Fe<sup>3+</sup> and SO<sub>4</sub><sup>2-</sup> on CH<sub>4</sub> emission in CWs. For instance, the application of 10 tons of gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O) per hectare to the sediment core of a FWS CW treating farm wastewater in Scotland reduced CH<sub>4</sub> emissions by 28%, whereas the addition of 5 tons of ochre (Fe(OH)<sub>3</sub> and FeO(OH)) ha<sup>-1</sup> caused methane emission to decrease by 63% (Pangala et al., 2009).

#### 4.5. The role of hydrological regime and plants

The impact of hydrological regime (pulsing hydrology) and aquatic macrophytes seems to be more problematic, and different studies sometimes yield contradictory results (Table 9).

In all cases, however, the pulsing hydrological regime in FWS CWs and sometimes HSSF CWs, and intermittent loading in VSSF CWs clearly decrease methane emission. Regarding N<sub>2</sub>O emission, some studies (Jia et al., 2011; Mander et al., 2011) show enhanced N<sub>2</sub>O release from surface flow CWs due to pulsing hydrology. A similar effect was shown by Van de Riet et al. (2013) for rewetted peatlands. Some other studies on artificial riverine wetlands, however, suggest that pulsing hydrology slightly decreases N<sub>2</sub>O emission (Hernandez and Mitsch, 2006).

Based on stepwise multiple regression analysis, we found a significant negative correlation between the hydraulic load (Q; Table 7) and CH<sub>4</sub> emission in HSSF CWs, however, suggesting an increase in the hydraulic load would decrease the water purification efficiency of the system, and the benefit of lower CH<sub>4</sub> release would be diminished.

Several studies show that extensive aquatic macrophyte cover significantly suppresses CH<sub>4</sub> emission in FWS CWs (Stadmark and Leonardson, 2005; Thiere et al., 2011; Mander et al., 2013) and artificial riverine wetlands (Altor and Mitsch, 2006; Sha et al., 2011). Hernandez and Mitsch (2006, 2007), Pulou (2011), and Thiere et al. (2011) have also shown the decreasing effect of aquatic macrophyte cover on N<sub>2</sub>O emission from FWS wetlands. However, considering all of the aquatic macrophytes individually, the effect on CH<sub>4</sub> and N<sub>2</sub>O emission can vary from decreasing to increasing. Some aerophytomatous macrophytes such as common reed (*P. australis*), which facilitate gas transport from the sediment (CH<sub>4</sub>) and to the sediment (O<sub>2</sub>), can inhibit CH<sub>4</sub> emission from wetlands (Brix, 1990, 1997; Ström et al., 2005). Likewise, oxygenation and the related methanogenesis-inhibiting effect have been demonstrated in an experimental landfill-leachate-treatment wetland with willow plants (Williams et al., 2010). On the other hand, several aerophytomatous wetland plants such as *Eriophorum vaginatum* (Waddington et al., 1996; Ström et al., 2005), *Juncus effusus* (Smialek et al., 2006; Schafer et al., 2012), *Typha latifolia* and *Zizania latifolia* (Inamori et al., 2007; Wang et al., 2008a,b) are important emitters of methane. Also, in some conditions *P. australis* can emit CH<sub>4</sub> (Juutinen et al., 2003; Duan et al., 2006).

Different wetland macrophytes show a remarkable variety in the results of N<sub>2</sub>O emission. Due to N uptake, the N<sub>2</sub>O emission in plant-covered CWs can be decreased (Silvan et al., 2005; Wang et al., 2008a,b; Maltais-Landry et al., 2009a,b). In contrast, Rückauf et al. (2004) showed in lab experiments that *Phragmites* plants significantly enhanced N<sub>2</sub>O emission, whereas *Phalaris arundinacea* did not affect N<sub>2</sub>O emissions, and no emission via the shoots was observed. Most probably, *Phragmites* plants were supplying more oxygen into the soil than the *Phalaris*.

The harvesting of *Phragmites* stands had a short-term decreasing effect on CH<sub>4</sub> emission from FWS CWs, but harvesting in HSSF CWs did not impact GHG emissions (Zhu et al., 2007). Nevertheless, following the multiple ecosystem services concept (Mitsch and Gosselink, 2007), FWS CWs can be used for both water quality improvement and plant biomass production that does not generate GHG emissions. However, further long-term studies in the field of the macrophytes' effect and their possible use for multiple purposes are needed (see Meerburg et al., 2010).

The relationship between algae growth and GHG emissions in treatment wetlands has not been thoroughly studied. Our study shows that in FWS CWs for domestic wastewater treatment, beds with filamentous algae of *Spirogyra* spp. emitted more N<sub>2</sub>O and less CH<sub>4</sub> than beds with aquatic macrophytes and non-vegetated beds (Tables 1 and 2). Anderson and Mitsch (2006) have found that in created riverine wetlands in deeper open water (OW) zones with sometimes intensive algae growth, mean sediment accumulation was significantly higher than that in zones of emergent vegetation. Large accumulations of Ca and inorganic C in the OW zones of

wetlands suggest that  $\text{CaCO}_3$  deposition has remained a critical process where algae productivity has been highest (Anderson and Mitsch, 2006). These data suggest that algae growth may be a significant facilitator of C sequestration in FWS CWs, although in the earlier stages of FWS CWs this can, however, can lead an increase in BOD in the outlet (Diaz et al., 2012).

Constructed wetlands accumulate OM over time (Mander et al., 2008, 2014; Van der Zaag et al., 2010), forming in FWS CWs thicker organic layers and causing oxygen deficiency, which facilitates methanogenesis and results in elevated  $\text{CH}_4$  emission. To mitigate the negative role in the earlier stages after wetland creation, support of aquatic macrophytes' cover and its controlled management is recommended in the further application of FWS and HSSF CWs.

#### 4.6. Significance of emission values found

The emission factors reported here are limited to the process emissions and therefore do not include the amount of GHG produced during the generation of electric power which is necessary, for example, for feeding batch wetlands though the use of pumps (particularly VSSF), artificial aeration in aerated wetlands, or for running activated sludge processes. The total operational carbon footprint of a treatment technology will be a combination of the process emissions (i.e., emission factors reported here) and the operational carbon associated with energy use, estimated at 0.544 kg  $\text{CO}_2$  per kWh used (UKWIR, 2008). The amount of energy required to run a particular treatment plant is dependent on many factors, including the type of treatment technology (i.e., biofilm vs. suspended growth), topography (i.e., gravity vs. pumped flows), efficiency of the instrumentation (i.e., size of the blower or pump), instrument run times (i.e., how many batch cycles are operated), among many others. Thus, analogous to the conventional energy benchmarking among treatment plants to determine process efficiency in terms of energy used (and its associated carbon footprint) and proposing optimization strategies, it is also possible to compare treatment efficiency in terms of mass of GHG emitted as process emissions generated from removing pollutants from wastewater (i.e., emission factors).

In comparison with conventional wastewater treatment systems, CWs emit significantly lower levels of  $\text{N}_2\text{O}$ . Czepliel et al. (1995) show that the total  $\text{N}_2\text{O}$  emissions from the municipal wastewater treatment systems in Durham, NH, USA were estimated to be  $6.6 \times 10^7 \text{ g of N}_2\text{O yr}^{-1}$  from primary treatment and  $1.2 \times 10^9 \text{ g of N}_2\text{O yr}^{-1}$  from secondary activated sludge treatment. The hourly rates of  $\text{N}_2\text{O}$  emission ranged from 5 to 75 mg  $\text{N}_2\text{O-N m}^{-2} \text{ h}^{-1}$  over all parts of the system (Czepliel et al., 1995), which is one to three magnitudes higher than the emission from wastewater treatment wetlands (Tables 2, 4 and 6). Likewise, Park et al. (2000) measured significantly higher  $\text{N}_2\text{O}$  emission values in a domestic wastewater treatment SBR than those found in CWs.

However, comparisons of CW emissions with energy intensive wastewater treatment systems (e.g., activated sludge plants) are possible by examining the emission factors and comparing those against systems with similar treatment goals. That is, comparison of hourly emissions per  $\text{m}^2$  is not the best way, as the volume of the treatment systems (i.e., 4–5 m aeration tanks vs. 0.6–1 m deep wetlands) and the area requirement per person equivalent are far too different for direct comparisons ( $0.05\text{--}0.1 \text{ m}^2 \text{ PE}^{-1}$  for activated sludge vs.  $3\text{--}5 \text{ m}^2 \text{ PE}^{-1}$  for CWs). In this context, the EF of VSSF systems should typically be compared against the EF of other nitrifying systems, and FWS and HSSF systems should be compared against denitrifying or TN removing systems.

The mean EF of 0.023% of  $\text{TN}_{\text{in}}$  from nitrifying VSSF wetlands is significantly lower than the 0.16–0.6% reported for biological

nutrient removal plants (Ahn et al., 2010), and the 0.036% from a nitrifying activated sludge plant (Aboobakar et al., 2013).

For HSSF wetlands, the mean emission factor of 0.79% of influent TN is within the range of 0.01–1.8% reported for energy intensive technologies (Ahn et al., 2010).

The life cycle assessment (LCA) in combination with GHG emission values shows the benefits of CWs. Pan et al. (2011) demonstrate that a conventional wastewater treatment plant system in China emits 7.3 kg  $\text{CO}_2$ -equivalents ( $\text{CO}_2\text{-eq}$ ) including both  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$  fluxes calculated based on radiative force coefficients; IPCC, 2007 to remove 1 kg BOD in the studied life cycle, while the VSSF CW system only emits 3.18 kg  $\text{CO}_2\text{-eq}$ . To a large extent, this is due to significantly lower  $\text{CH}_4$  emission in the studied VSSF CWs. Although the EF value of  $\text{CH}_4$  emissions in FWS CWs is high (Table 8) the final efficiency of these systems may even be higher due to the lower need for construction materials and energy than in VSSF CWs (Vymazal, 2001, 2007). Similarly, the LCA of subsurface flow CWs, which included GHG emission, showed that in comparison with HSSF CWs, the VSSF CWs are less impactful configuration for wastewater treatment, especially for removing TN from domestic wastewater (Fuchs et al., 2011).

However, the EF values for  $\text{N}_2\text{O}$  in all analyzed CW types and even in studied conventional wastewater treatment systems are lower than those found on agricultural soils in Great Britain, where annual EFs varied from 0.4% to 6.5% of the N applied (Dobbie and Smith, 2003), and were comparable with the present IPCC default EF for  $\text{N}_2\text{O}$  of 1.25% of the N applied (IPCC, 2006). Thus, the mitigation of GHG emissions from treatment systems should be tackled alongside mitigation strategies for emissions from agricultural activities.

#### 4.7. Further study perspectives

Among further investigations that would contribute to a better understanding of C and N cycling, the continuous measurement of  $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes using transparent automatic chamber method and eddy covariance technology can be highlighted. For VSSF and HSSF CWs with relatively small areas, the chamber method gives adequate estimates, although automatic and transparent chambers make it possible to fill the measurement gaps and analyze full C and N budgets (see, e.g., Chojnicki et al., 2010). In larger FWS CWs the eddy covariance method is preferable.

In order to distinguish between different sources of  $\text{N}_2\text{O}$  (denitrification vs. nitrification),  $\text{N}_2\text{O}$  isotopomer studies (Meijide et al., 2010; Well et al., 2012; Wunderlin et al., 2013) have yielded promising results and must be used in further investigations in wetlands. Some studies (Riya et al., 2010) suggest distinguishing between the direct and indirect GHG emissions in CWs, similarly to analogous studies on  $\text{N}_2\text{O}$  fluxes from agricultural landscapes (IPCC, 2007; Vilain et al., 2012) and global estimations of  $\text{N}_2\text{O}$  emissions (Seitzinger and Kroese, 1998). Riya et al. (2010) evaluated direct  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions (gas fluxes from the CW surface water to the atmosphere) and indirect  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions (dissolved gas concentrations in filtered water) from a VSSF CW planted with forage rice, and found that the percentages of indirect emission to total (direct + indirect) emission during the experimental period were 2.9% and 86.7% for  $\text{CH}_4\text{-C}$  and  $\text{N}_2\text{O-N}$ , respectively. Emission factor of the indirect  $\text{N}_2\text{O-N}$  emission was 0.053–0.86%. This is comparable to those of indirect emission sources in the literature and also to the results of our study. However, these results indicate the importance of monitoring and controlling indirect  $\text{N}_2\text{O-N}$  emission from CWs (see also Well et al., 2005, 2012). In terms of better performance and optimal maintenance of CWs, studies on the impact of hydrological regimes and the seasonal development

of vegetation and microbial communities on the emission of GHGs and on C and N budgets must be carried out.

Better understanding of the role of microorganisms in key processes influencing CH<sub>4</sub> and N<sub>2</sub>O emissions, methanogenesis and denitrification is another important challenge for further investigations. Analysis of the structure of microbial communities and functional gene abundance and diversity studies using next generation sequencing and quantitative PCR techniques are already widely used, and the first results for FWS CWs (Garcia-Lledo et al., 2011) and artificial riverine wetlands (Song et al., 2012; Ligi et al., 2014a,b) have been published.

## 5. Conclusions

The emission factor of CH<sub>4</sub> in FWS and HSSF CWs has been found to be very high, yet the absolute value of CH<sub>4</sub> emission is relatively small, and 1–2 magnitudes lower than that found in conventional wastewater treatment plants. Nevertheless, GHG emission is an important factor in the operation of CWs, but the main task is the optimization of water purification processes.

The use of peatlands in the creation of treatment wetlands has benefits regarding water quality improvement, but can significantly increase CH<sub>4</sub> emission. The overloading of HSSF CWs will lead to clogging and to elevated CH<sub>4</sub> emission, and the use of FWS CWs for raw wastewater treatment will also cause intensive OM accumulation but very high CH<sub>4</sub> emission.

To mitigate CH<sub>4</sub> emission from FWS and HSSF CWs, a pulsing hydrological regime (fluctuating water table and intermittent loading) is recommended. This can, however, elevate N<sub>2</sub>O emission, since the global warming potential (GWP) of these systems is mainly determined by CH<sub>4</sub> emissions, and the intermittent loading and pulsing hydrology can be practical. Likewise, support for the development of aquatic macrophyte cover and controlled harvesting in FWS and HSSF CWs can help minimize GHG emissions.

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