

1 **Carbon and Nitrogen Dynamics and Greenhouse Gases Emissions in Constructed**
2 **Wetlands Treating Wastewaters: A Review**

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34 **Abstract**

35 Globally, constructed wetlands (CWs) are a developing technology for the removal of pollutants
36 (e.g. carbon- C and nitrogen- N) from wastewater. The reported removal efficiency of C and N in
37 CWs is very inconsistent and does not in itself reveal whether the removal processes are due to
38 physical attenuation or whether the different species have been transformed to other reactive
39 forms. The existing international literature could not provide an explanation for such low removal
40 efficiencies and typically does not consider the fate of the removed nutrients. A comprehensive
41 review is therefore necessary to assist with the understanding of removal mechanisms and
42 process kinetics of C and N in CWs, which underpin their environmental benefits. Herein, we
43 provide an overview of the current knowledge and discussion concerning the biogeochemical
44 processes that control nutrient removal in CWs used for treating wastewater and the likely
45 impacts of these processes on aquatic and atmospheric environments. This paper does not only
46 identifies knowledge gaps that currently inhibit a holistic understanding of the fate of various C
47 and N species but also itemises methodologies to elucidate the necessary data to eliminate such
48 knowledge gaps. It highlights that nutrient removal efficiencies in CWs vary with CW type,
49 vegetation type, climate, season, treatment region and other management practices. No single CW
50 type shows an optimum performance in removing C and N pollutants. Horizontal flow CWs tend
51 to show higher performance for nitrate (NO_3^-) removal as they provide good conditions for
52 denitrification but cannot remove ammonium (NH_4^+) due to limited ability to nitrify NH_4^+ .
53 Vertical flow CWs show higher performance for NH_4^+ removal but their denitrification ability is
54 low. Surface flow CWs decrease N_2O emissions but increase CH_4 emissions; subsurface flow
55 CWs increase N_2O and CO_2 emissions but decrease CH_4 emissions. Mixed species perform better
56 than monocultures in increasing C and N removal and decreasing GHG emissions but
57 experimental evidence is still scarce. Lower hydraulic loading with higher hydraulic retention
58 time enhances nutrient removal but more experimental evidence is required to determine an
59 optimum design. A conceptual model is presented which highlights the current state of
60 knowledge and suggests specific experimental work across CW, vegetation and wastewater type,
61 hydraulic loading rate and retention time and hydrologic pulsing headings. We recommend that
62 further research on process based C and N removal and on the balancing of end products into
63 reactive and benign forms is critical to the assessment of the environmental performance of CWs.

64

65 Keywords: Carbon, nitrogen, constructed wetlands, pollution swapping, nitrous oxide, methane

66

67 1. Introduction

68 Increasing anthropogenic loading of reactive nitrogen (Nr; all forms of nitrogen except di-
69 nitrogen gas, N₂) along the nitrogen (N) cascade in the environment raises many critical concerns
70 for human health, drinking water quality (Gray, 2008), coastal and marine water degradation
71 causing eutrophication, as well as algal blooms and hypoxia (Conley et al., 2009; Rabalais et al.,
72 2010). There are natural sinks for Nr along the N cascade (Galloway et al, 2003; Tanner et al.,
73 2005) but other sinks that are rehabilitated or artificial, may also be introduced and evaluated at
74 key locations e.g. constructed wetlands (CWs) (Gold et al., 2013) or permeable reactive
75 interceptors (Fenton et al., 2014). This review focuses on CWs treating wastewater which are
76 now an emerging technology used globally as a mitigation option for nutrient retention at the
77 delivery end of the transfer continuum (Dunne, et al., 2005; Sjøvik et al., 2006; Tanner and
78 Kadlec, 2013; Tanner and Sukias, 2011). Although CWs have a proven potential for the removal
79 of organic carbon (C) and N, studies have shown that removal efficiencies have been highly
80 variable, due to inadequate observations of C and N transformations and their removal
81 mechanisms (Seitzinger et al., 2002) and lack of improved adoption and placement (Gold et al.,
82 2013). Constructed wetlands are complex bioreactors which facilitate a number of physical,
83 chemical and biological processes but often continue to be evaluated as a 'black box' in terms of
84 process understanding (Langergraber, 2008). Many investigations target single contaminant
85 remediation, whilst disregarding the reality of mixed contaminants entering and leaving such
86 systems. They do not consider the dynamic of pollution swapping (the increase in one pollutant
87 as a result of a measure introduced to reduce a different pollutant) driven by transformational
88 processes within and around the system. The CWs can have negative climatic impacts (Ström et
89 al., 2007), act as an active source of greenhouse gas (GHG) (IPCC, 2014; Clair et al., 2002;
90 Mander et al., 2008; Mitsch and Gosselink, 2000) and enhance pollution swapping (Reay, 2004).
91 There are many pathways by which the removed N can contribute to water and air pollution:
92 accumulation and adsorption in soils, leaching to groundwater (nitrate-NO₃⁻ and ammonium-
93 NH₄⁺), emission to atmosphere via nitrous oxide- N₂O and ammonia and/or conversion to di-
94 nitrogen (N₂) gas. The kinetics of these pathways/processes varies with CW types and
95 management practices and therefore needs to be studied quantitatively for their sustainable use.
96 The impacts of these factors on C and N cycling and associated GHG emissions, especially in
97 response to a changing climate, are poorly known.

98

99 Several processes can produce N₂O in CWs: nitrification, denitrification and nitrifier
100 denitrification. It is known that CWs significantly contribute to atmospheric N₂O emissions either
101 directly to the atmosphere from the surface of the wetland as its by-product (IPCC, 2014; Søvik
102 et al., 2006; Ström et al., 2007; Elberling et al., 20011; Van der Zaag et al., 2010) or indirectly via
103 dissolved N₂O in the effluent or groundwater upon discharge to surface waters (Riya et al., 2010).
104 The global importance of this N₂O emission has been increasingly recognised (IPCC, 2014).
105 Indirect N₂O emissions from CWs effluents that discharge directly to aquatic environments have
106 a default emission factor (EF) of 0.005 but with a wide range (0.0005-0.25). The wide range of
107 EF is highlighting the large uncertainty, which warrants further refinement.

108
109 Constructed wetlands receive organic C from the wastewater, being used for treatment, and also
110 from fixation by the photosynthetic hydrophytes which are rapidly recycled by respiration but
111 much of this is incorporated into soil as organic C. Soil organic C undergoes the biogeochemical
112 processes that regulate C accretion in soil and microbial respiration, producing carbon dioxide
113 (CO₂). Anaerobic mineralization of organic C by methanogenic archaea can produce methane
114 (CH₄) and exchange with atmosphere (Laanbroek, 2010; Ström et al., 2007; Søvik et al., 2006;
115 Pangala et al., 2010). Constructed wetlands can contribute to the dissolved organic carbon (DOC)
116 load transfer to ground and surface waters which can produce and exchange substantial amounts
117 of CO₂ and CH₄ with atmosphere (Clair et al., 2002; Elberling et al., 2011). The CWs are thus
118 can diminish the environmental benefits of wastewater treatment in the exchange of GHG which
119 need to evaluate. The dynamics of dissolved N₂O, CO₂ and CH₄ in CWs is a key “loop hole” in
120 global greenhouse gas budgets.

121
122 Surface emissions of GHG from CWs have been commonly measured by closed chamber method
123 (Johansson et al., 2003, 2004; Mander et al., 2005, 2008) but rarely measured by ebullition and
124 diffusion methods (Søvik et al., 2006). The measured rates have shown high spatial, temporal and
125 diurnal variations due to the change in biogeochemistry of C and N and plant-microbes-soil
126 interaction over time and space. The surface emissions cannot explain the kinetics of production
127 and consumption rates of GHG which we need to know for adoption of better management to
128 mitigate emissions. In addition, subsurface export of dissolved nutrients and GHG, being an
129 important pathway of nutrient loss (Riya et al., 2010), is ignored. Mass balance analysis of the
130 different components of the N cycle and kinetics of their transformation processes occurring
131 within the treatment cells using the isotope-tracing ¹⁵N technique can provide mechanistic
132 information for N transformation products (Lee et al., 2009; O’Luanaigh et al., 2010) and

133 therefore can be used to start to answer such questions. Similarly, ^{14}C application and
134 measurement of C species (e.g. CO_2 , CH_4 , and DOC) with the soil core incubation could
135 elucidate the C mineralization and CO_2 and CH_4 production and consumption. These methods in
136 combination will be able to provide a comparative analysis of the rates of C and N transformation
137 processes and role of these processes in delivering NO_3^- and or NH_4^+ and DOC to ground/surface
138 waters and N_2O , CO_2 and CH_4 to atmosphere. Past reviews on CWs, though very limited,
139 summarise the performance of different types of CWs on C and N removal (Vymazal, 2007) and
140 surface emissions of GHG (Mander et al., 2014) but have not discussed the mechanisms of
141 nutrient removal and the fate of the nutrients delivered and removed to and from CWs. The
142 objectives of this review are i) to understand the biogeochemical dynamics of C and N in CWs ii)
143 to better understand the fate of various C and N species in a holistic manner, in addition to the
144 conventional influent – effluent balance for nutrient removal iii) to identify the research gaps that
145 need to be undertaken for optimising nutrient removal efficiencies and mitigating GHG emissions
146 and iv) to focus on the advanced methods that can give insights into the understanding of
147 production and reduction of GHG.

148

149 **2. Removal Efficiency, Hydraulic Loading and Retention Time**

150 In CWs, the efficiency of C and N removal is generally limited and highly variable over CW
151 types, plant types, seasons, climatic regions and management practices. On average, it appears
152 that 50 and 56% of the influent TN and TOC, can be removed respectively, but the removal rates
153 are very inconsistent. Mean ($\pm\text{SE}$) TN removal efficiencies, obtained from the literature cited,
154 ranged from $31.3 \pm 6.3\%$ in surface flow (SF) CWs to $40.4 \pm 4.4\%$ in subsurface flow CWs
155 whereas TOC removal ranged from $18.8 \pm 9.4\%$ in SF CWs to $56.2 \pm 9.5\%$ in vertical subsurface
156 flow CWs (Table 1 and Table 2). In European systems, for example, typical removal percentages
157 of ammoniacal-N in long-term operation are c. 35% but can be enhanced if some pre-treatment
158 procedures are followed (Verhoeven, and Meuleman, 1999; Luederitz et al., 2001). Generally,
159 TN removal is higher in surface flow (SF) CWs than subsurface flow (SSF) CWs (Table 1) but
160 there is no straight forward relationship observed in previous studies. For example, Van der Zaag
161 et al. (2010) showed higher N removal in SF CWs than SSF but Søvik et al. (2006) and Gui et al.
162 (2007) showed the opposite. In SSF CWs, limited removal can be caused by the typically reduced
163 environment that enhances NH_4^+ accumulation and limits NH_4^+ oxidation. In SF CWs,
164 denitrification rates can be limited due to lack of NO_3^- . In vertical subsurface flow (VSSF) CWs,
165 aeration can increase NH_4^+ oxidation to NO_3^- which can be denitrified or converted to NH_4^+ by
166 DNRA. Plant species are important structural components of CWs, which affect C and N

167 **removal.** Species selection based on the published data is still complex because some species are
168 efficient in removing one pollutant but not the other (Bachand and Horne, 2000; Bojcevska and
169 Tonderski, 2007; da Motta Marques et al., 2000). In some studies there are no inter species
170 differences at all (Calheiros et al., 2007). Impact of plant species on nutrient removal has not
171 been reported clearly in the published literature, where species differences for one or more
172 nutrient was observed, because of the problems with plant growth due to unpredicted
173 environmental conditions, wastewater toxicity, and in many cases the reasons were not specified
174 (Fraser et ., 2004; Solano et al., 2004; Haule et al., 2002). **Plants regulate CWs hydrology**
175 **(evaporation and transpiration), temperature (insulating CWs from seasonal temperature change,**
176 **trapping falling and drifting snow and heat loss of wind).** Mixed species perform better than
177 monocultures to remove C and N pollutants because they increase microbial biomass and
178 diversity. Some species can create large surface area for microbial attachment and enhance
179 microbial diversity, but experimental evidences are still scarce. Future research should investigate
180 the mechanisms of nutrient removal rather than the only empirical comparison in removal
181 efficiency for the plant species (Brisson and Chazarenc, 2009).

182
183 **Soil physicochemical properties are important factors controlling the purification capacity in**
184 **CWs. Soils with high permeability enhance downward nutrient movement to groundwater. High**
185 **cation exchange materials in soil enhance NH_4^+ fixation by soil matrix. Microbial activities and**
186 **growth depend on substrate C quality and C: N ratios which consequently affect nutrient removal**
187 **efficiency. Better growth of heterotrophic microorganisms is a function of the wastewater C: N**
188 **stoichiometry (Makino et al., 2003). A specific level of C:N ratio can favour one group of**
189 **microbes but disfavour some other groups. For example, higher C:N ratio can enhance**
190 **denitrification by providing electron donor for denitrifiers but the opposite can increase**
191 **nitrification. High C:N ratios can also encourage DNRA over denitrification. Yan et al. (2012)**
192 **showed the highest TN removal at C:N ratio 2.5: 1 coupled with the lowest TOC removal which**
193 **indicates that removal of one nutrient might lead to a problem to a different one. Future research**
194 **should therefore orient to maintain an optimum C:N ratio for achieving better water quality**
195 **standard. The uncertainty in removal efficiencies suggest that the rates of C and N**
196 **transformations and the fate of the removed nutrients within the CWs should be investigated.**
197 However, to our knowledge, no study is available that provides a holistic evaluation of C and N
198 attenuation and transformations.

199

200 The removal efficiency of pollutants in CWs depends on hydraulic loading rates (HLR) and
201 hydraulic retention time (HRT) (Toet et al., 2005). The HLR and HRT are considered to be
202 significant design parameters determining the nutrient removal efficiencies (Weerakoon et al.,
203 2013). Longer residence of wastewater in CWs increases the removal of C and N (Wang et al.,
204 2014) by increasing sedimentation and duration of contact between nutrients and the CW
205 systems. The effects of HLR and HRT can vary with the nature of the use of CWs e.g. whether
206 they are used for treating single or mixed pollutants. To reduce Nr delivery to the receiving
207 waters or to the atmosphere, CWs need to be optimally designed with respect to HLR and HRT.

208

209 Fluctuating hydraulic loading influences all biotic and abiotic processes in CWs. For example, if
210 the groundwater table is lowered through changes in hydraulic loading, soil aeration can increase
211 or decrease. Ammonification and nitrification rates increase with increased soil aeration and this
212 enhances C utilization by bacteria and therefore can stimulate the removal of TC and TN.
213 Investigation into the effects of fluctuating hydraulic loadings (hydraulic pulsing) on C and N
214 removal efficiencies and their transformation products will provide information about the fate of
215 the added nutrients in terms of their environmental benefits and or pollution swapping. For
216 example, if the dominant product is N₂, the system will be more benign to the environment but if
217 it is NH₄⁺, it can be fixed in the soils or transport to ground and surface waters connected to CWs
218 if the cation exchange sites become saturated. However, contrasting results on the impacts of
219 hydraulic loadings on nutrients removal efficiency are available in literature (Toet et al., 2005).
220 Luo et al. (2005) reported that low HLR results in incomplete denitrification, whereas Zhang et
221 al. (2006) argued that low HLR increases NH₄⁺ and chemical oxygen demand (COD) oxidation.
222 Song et al. (2011) reported an average HRT of 2.4 days during wetland operation periods for the
223 removal of NO₃⁻ and NH₄⁺ without documented design optima or understanding of their fate. The
224 way in which a CW performance is assessed can lead to different conclusions regarding removal
225 of Nr. For future studies, evaluation of systems in a holistic manner, which include pollution
226 swapping at different HLR and HRT is important, particularly within the context of the changing
227 hydrologic cycle in a changing climate. Also local legislative targets should be considered and
228 weighting factors (are GHG more important than water quality targets?) developed for different
229 species within the system to come up with overall balances for the CW. In addition to the
230 estimation of nutrient removal rates, investigation of the effect of HLR and HRT on the different
231 forms of nutrients in the final effluents and their fate in the natural environment can attempt to
232 elucidate the pollution swapping potential of CWs.

233

234 3. Accumulation of C and N in CWs Soils

235 The soil in CWs is a major sink for C and N (Mustafa and Scholz, 2011). Data on the C and N
236 accumulation (dissolved/particulate organic carbon - DOC/POC (labile or stable), **dissolved**
237 **organic N**- DON, TN, NH_4^+ or NO_3^- -N) within the soil profile of various CWs are scarce as most
238 studies focus on N balances between influent and effluent N loads. **A wide range of N**
239 **accumulation has been reported in the literature (e.g. 30-40%, Shamir et al., 2001; 39%,**
240 **Harrington et al., 2007; 9%, Mander et al., 2008; 2.5%, Obarska-Pempkowiak and Gajewska,**
241 **2003). This wide range could be due to the variations in CW types and management strategies.**
242 **The accumulated species of N are reactive unless they have been transformed to N_2 by**
243 **biogeochemical processes.** However, there is a dearth of information on the extent of Nr
244 accumulation in soils and discharge to surface waters and air (Shamir et al., 2001). **Accumulated**
245 **organic N could be mineralised to NH_4^+ and NO_3^- depending on the physico-chemical properties**
246 **of soil. The Nr could be assimilated by plants and microbes which are recycled in a soil-plant-soil**
247 **continuum. Nitrogen spiralling occurs from NH_4^+ to organic N and back to NH_4^+ within the CW**
248 **(O’Luanaigh et al., 2010). Typically, N accumulation has been found to decrease with soil depth**
249 **(Shamir et al., 2001). In terms of the conventional input-output balance, these are considered as**
250 **removed N, but in fact in such a biogeochemically active system they still remain. In addition to**
251 **N, organic C accumulation occurs in CW soils (Nguyen, 2000). As such, soils of CWs represent**
252 **organic C and Nr rich systems where the products of the continuously occurring biogeochemical**
253 **processes can be transported to fresh waters and to the atmosphere. Estimation of the rates of**
254 **nutrient accumulation in soils in various types of CWs under different management systems is**
255 **important. The stability of the accumulated C and N under changing climatic scenarios also needs**
256 **to be addressed to consider the long term sustainability of CWs.**

257

258 4. C and N Dynamics and Greenhouse Gas Emissions

259 Increased nutrients input to the CWs increases the productivity of wetland ecosystems and thus
260 increase the production of GHG. **As CWs are designed to remove pollutants in an**
261 **anaerobic/suboxic environment, they change the C and N biogeochemistry and contribute**
262 **significantly to CH_4 and N_2O emissions (Johansson et al., 2002, 2003; Mander et al., 2005, 2008;**
263 **Stadmark and Leonardson, 2005; Liikanen et al., 2006). Søvic et al. (2006) measured N_2O , CH_4**
264 **and CO_2 emissions in various CWs in different European countries and suggested that the**
265 **potential atmospheric impacts of CWs should be examined as their development is increasing**
266 **globally. Management of CWs must consider the negative climatic aspects of increased emissions**

267 of GHG in addition to their primary functions (Ström et al., 2007). Therefore, estimation of the
268 contribution of CWs to global warming is required. In this regard, measurement of spatial and
269 temporal variations (seasonal and diurnal) of GHG emissions is required to accurately estimate
270 CWs derived GHG emissions. A holistic assessment of ecologically engineered systems has been
271 thoroughly outlined in Healy et al. (2011, 2014) and developed further in Fenton et al. (2014).
272 Such assessments can be applied in evaluating nutrient dynamics in CWs. Moreover, plant
273 mediated GHG emissions could be an important component of total emissions but again research
274 in this area is very limited. Effective modelling or up-scaling of GHG emissions from watershed
275 to regional/ national scales is important for the improvement of global GHG budgets. Such up-
276 scaling needs an accurate estimation of C and N inputs and outputs i.e. a balance coupled with net
277 GHG emissions while considering all possible processes and pathways involved. So, studying the
278 dynamics of C and N in CW systems is crucial as the forms of removed C and N are particularly
279 pertinent to their potential for pollution swapping; global warming and water pollution.

280

281 Processes involved in N removal and N transformations in wetlands include sedimentation of
282 particulates (Koskiaho, 2003); nitrification, denitrification and DNRA (Poach et al., 2003; Burgin
283 et al., 2013), microbial assimilation and plant uptake-release (Findlay et al., 2003), **anammox and**
284 **deamox (DENitrifying AMmonium OXidation)**. Constructed wetlands are a complex system
285 which facilitates aerobic and anaerobic microsites. This means that nitrification and
286 denitrification can occur simultaneously. Nitrification, denitrification and nitrifier denitrification
287 are the processes responsible for the production of N₂O. Depending on the environmental
288 conditions or management practices prevailing, a certain process will dominate e.g.
289 denitrification is the dominant process in SF CWs (Beaulieu et al., 2011) but nitrifier
290 denitrification is dominant in VSSF CWs (Wunderlin et al., 2013). Generally CWs are anaerobic
291 but aquatic macrophytes can transport oxygen from the atmosphere to the rooting zone where it
292 can sustain nitrification. Nitrate produced in the rooting zone can be taken up by plants or
293 denitrified and/or converted back to NH₄⁺ by DNRA. The DNRA is an anaerobic process where
294 NO₃⁻ is transformed to NH₄⁺ which can remain in that form only until it has contact with an
295 aerobic environment (Tesoriero et al., 2000), after which it is oxidized to NO₃⁻ (Thayalakumaran
296 et al., 2008). The fermentative bacteria which carry out DNRA are obligate anaerobes (Hill,
297 1996) and so cannot occupy all the niches that denitrifiers can. Takaya (2002) stated that more
298 reducing state favours DNRA over denitrification. However, the conditions that favour the
299 occurrence of either denitrification or DNRA are still in debate (Rütting et al., 2011). The
300 differences between denitrification and DNRA may be due to the availability of organic matter,

301 because DNRA is favoured at high C:N ratio. The N_2O produced in soil can be consumed by
302 denitrifiers and/or emitted to the atmosphere by ebullition and diffusion processes or via vascular
303 plants. Denitrification has been estimated to be a significant N removal process but actual
304 quantification data are scarce. Limited studies have estimated N losses by denitrification e.g. 19%
305 (Mander et al., 2008) and 86% (Obarska-Pempkowiak and Gajewska, 2003) of the total N input
306 based on the mass balance study. To our knowledge, no data are available on denitrification
307 measurements in soil/subsoils of surface flow CWs. While many of these pathways transfer Nr
308 (mainly NH_4^+ and N_2O) to the environment, other pathways can convert Nr to N_2 (e.g.
309 denitrification, anammox and deamox). Anammox can remove NO_2^- and NH_4^+ as N_2 when the
310 existing environment is hypoxic. The deamox can remove NO_3^- and NH_4^+ as N_2 where NO_3^- is
311 converted to NO_2^- by autotrophic denitrification with sulphide (Kalyuznyi et al., 2006). In CWs,
312 anammox and DEAMOX are not well understood and so it is crucial to identify which of the
313 processes are occurring in a specific type of CWs and at what rate they occur. Once a process that
314 provides N_2 as the end product is determined in a specific system then the CW management could
315 be directed towards enhancement of that process. Hence, quantifying the rates of these processes
316 under various CW types is required for improved N management towards lowering Nr in the
317 environment.

318
319 The various components of the C cycle include: fixation of C by photosynthesis, respiration,
320 fermentation, methanogenesis and CH_4 oxidation with reduction of S, Fe and NO_3^- . Anaerobic
321 methane oxidation coupled with denitrification, a recently proposed pathway of the C cycle (á
322 Norði and Thamdrup, 2014; Haroon et al., 2013; Islas-Lima et al., 2004), can reduce CH_4
323 emissions in CWs. The C removal processes are sedimentation, microbial assimilation, gaseous
324 emissions, dissolved C losses through water to ground and surface water bodies and chemical
325 fixation (bonding with chemical ions). Net primary productivity (NPP) of wetland hydrophytes
326 varies across CW type, season, climatic region and local environmental conditions. For example
327 taking the same plant species results can vary remarkably in different geographical regions (Brix
328 et al., 2001). Carbon mineralization in sediments depends on the redox chemistry of soil,
329 availability and quality of C (labile or recalcitrant) and temperature. In CWs, C cycling is very
330 complex due to the changes in redox chemistry which regulates production and consumption of
331 CO_2 and CH_4 (Brix et al., 2001). In low redox conditions with limited dissolved oxygen,
332 methanogens can consume DOC and thus it is conducive to CH_4 production. The C:N ratios of
333 wastewater affect microbial growth and development which in turn affect their response to C and
334 N cycles and GHG emissions. Previous research on the effects of C:N ratios on nutrient removal

335 and GHG emissions are limited. A few examples include Yan et al. (2012) and Zhao et al. (2014),
336 which showed lower CO₂ and CH₄ emissions at C:N ratios between 2.5:1 and 5:1, but this lower
337 range of C:N ratios decreased TOC removal. Hence, investigation of the influence of C:N ratio
338 on nutrient removal efficiencies and GHG emissions across CW and management types is crucial.
339 In summer, oxygen diffusion to the topsoil can reduce methanogenesis and stimulate CH₄
340 oxidation (Grünfeld and Brix, 1999). However an increase in temperature can decrease dissolved
341 oxygen in deeper subsoil layers, which can enhance CH₄ production. Only a limited number of
342 studies have considered CH₄ and CO₂ efflux from CWs (e.g. Mander et al., 2008). Like for all
343 biochemical reactions temperature, increases C and N turnover in CWs causing high variations in
344 GHG emissions under same plant species in different regions (temperate/ tropical/ arctic). These
345 variations need to be considered while extrapolating GHG emissions for different types of CWs
346 under different management practices.

347
348 Emissions of GHG in CWs can vary across CW typologies e.g. surface flow or subsurface flow
349 (Van der Zaag et al., 2010). A summary of N₂O, and CO₂ and CH₄ were presented in Table 3 and
350 Table 4, respectively. Generally CH₄ emissions are higher in SF CWs than the SSF CWs (Table
351 3) but vary with seasons. Nitrous oxide and CO₂ emissions are higher in VSSF CWs than HSSF
352 and SF CWs. The N₂O emissions factors (EF; N₂O/TN input*100) ranged from 0.61 ± 0.21% in
353 SF CWs to 1.01 ± 0.48% in VSSF CWs. The EF for CH₄ emissions ranged from 1.27 ± 0.31% in
354 VSSF CWs to 16.8 ± 3.8% in SF CWs. The GHG from CWs can vary between vegetated and
355 non-vegetated systems (Table 5). Aquatic plants play important role on GHG production (provide
356 C necessary for microbes) and transport to the atmosphere by releasing GHG through their
357 interconnected internal gas lacunas (Laanbroek, 2010). Emergent plants can transport
358 atmospheric oxygen to the rooting zone and contribute to increased N₂O and CO₂ production and
359 CH₄ consumption (Brix, 1997). Vascular plants can exchange GHG between the rooting zone and
360 atmosphere (Yavitt and Knapp, 1998). Vegetation and its composition affect the nutrient
361 dynamics and the production, consumption and transport of greenhouse gases and hence their
362 exchange between wetlands and atmosphere (Ström et al., 2003, 2005; Søvic et al., 2006;
363 Johansson et al., 2003). They can also affect the biogeochemistry of CWs due to the differences
364 in their growth and development, longevity, root systems, root density, root depth and microbial
365 ecology in the rhizosphere. Some plant litter as it decomposes can release organic matter with
366 lignocellulose and humic compounds that are more or less labile or stable in nature than others.
367 Release of low molecular weight organic matter that is labile in nature is more likely to produce
368 GHGs than stable forms. For example, *Z. latifolia* showed higher nutrient removal and CH₄

369 fluxes than *P. australis* (Inamori et al., 2007). *Z. lotifolia* root system is shallow and the activity
370 of methanotrophs was primarily confined to the top soil. The root systems of *P. australis* are
371 deeper which is more favourable to oxidise CH₄. Fluctuating water table in CWs has significant
372 impacts on GHG dynamics. Pulsing hydrologic regimes decreases CH₄ but increases N₂O
373 emissions. In aerobic and anaerobic conditions caused by pulsing hydrology, incomplete
374 nitrification and denitrification increase N₂O emissions. However, the effects of pulsing
375 hydrologic regimes on GHG emissions are contradictory. For example, intermittent hydrologic
376 regimes decrease both N₂O (Sha et al., 2011) and CH₄ emissions (Song et al., 2010). Highly
377 contrasting results on gas emissions with fluctuating water levels have been reported and the
378 controlling mechanisms are unclear (Elberling et al., 2011). Therefore, the assessment of GHG
379 emissions in various types of CW (surface flow, subsurface flow; vertical and horizontal),
380 vegetation used (vegetated, nonvegetated, plant species composition) and under different
381 management systems (HLR, HRT, soil used and water table) is necessary in light of the national
382 and global GHG budgets and mitigation of GHG emissions. In addition, such measurements will
383 help scientists, environmental managers and policy makers to adopt environmental friendly
384 construction and management of CWs. The enhanced reduction of N₂O to N₂ needs further
385 elucidation. Assessment of the reactive versus the benign forms of C and N transformation
386 products in various types of CWs will give insights into their environmental friendly design and
387 management.

388

389 5. Surface Emissions vs Subsurface Export of C and N

390 Dissolved GHG produced in soils and subsoils can be emitted to atmosphere by transpiration of
391 vascular plants (from within the rooting zone), ebullition and diffusion from soils. Elberling et al.
392 (2011) reported that in wetlands, the transport of gases through subsoil occurs both via diffusive
393 transport in the pores and through the vascular plants. Surface emissions of GHG from CWs are
394 well recognised and have been commonly measured by chamber methods. The GHG produced in
395 CWs can also be transported to the groundwater with the percolating water and emit to
396 atmosphere upon discharge to surface waters. It can also flow towards surface waters by
397 advective transport and or by dispersion of groundwater. Dissolved nutrients can be preferentially
398 leached down into deeper soil layers and groundwater via different pathways (e.g. root channels).
399 The Nr delivered to groundwater can be transformed *in situ* to other reactive or benign forms.
400 Hence, quantification of such Nr loadings to groundwater and their *in situ* consumption (e.g. N₂O
401 to N₂ or CH₄ to CO₂) is required to understand their environmental consequences. In addition,
402 DON, NO₃⁻ and NH₄⁺ and DOC delivered to surface waters can undergo biochemical reactions

403 and produce N₂O, CO₂ and CH₄ in streams and estuaries. Ström et al. (2007) measured a
404 considerable quantity of CH₄ in porewater and found a correlation between the surface emissions
405 and porewater CH₄ concentrations in vegetated wetlands. Measuring only the surface emissions
406 of GHG can miss substantial quantity of GHG emissions from CWs. For example, Riya et al.
407 (2010) measured indirect emissions of CH₄ and N₂O accounting for 2.9 and 87% of total
408 emissions. Measuring porewater GHG and linking these to the surface emissions and subsurface
409 export to groundwater below CWs will help to estimate a better GHG balance from both a
410 national and global context. Elberling et al. (2011) linked subsurface gas concentrations in
411 wetlands to the surface fluxes using a diffusion model which has demonstrated the need for future
412 studies on subsurface GHG production, consumption and net GHG emissions in CWs ecosystem
413 in a climate change context. It is important to characterise soils and subsoils physical (e.g.
414 texture, bulk density) and hydraulic (development of a soil water characteristic curve) properties
415 and to assess their potential to percolate dissolved nutrients and gases in the solute phase to the
416 underlying groundwater. To our knowledge, indirect pathway of GHG emissions from CWs has
417 never been reported despite the fact that this would appear to have a high biogeochemical
418 potential to produce and exchange GHG. The balance between N and C input and output flows
419 between CWs and aquatic and atmospheric environments together with the direct and indirect
420 emissions of C and N species could be an important input to global C and N budgets.

421

422 6. Hydrogeochemistry below CWs

423 The CWs can be designed with or without a clay liner or a compacted soil bed at the base, which
424 can lead to large differences in permeability of the underlying layers. The variation in
425 permeability of a CW soil bed will affect solute, nutrient and GHG flows and their interactions
426 with the underlying groundwater (Dzakpasu et al., 2012; 2014). Groundwater hydrogeochemistry
427 below CWs can therefore provide a unique insight into such interactions. An example of such
428 interactions would be between nutrient rich water discharging from CW cells mixing with
429 laterally moving regional groundwater. It should be noted that groundwater can also discharge
430 into CWs depending on the hydraulic gradients. This necessitates that fully screened multiple
431 multi-level piezometers or boreholes are installed at such sites to elucidate groundwater flow
432 direction, hydraulic gradients and hydraulic permeability. Such monitoring networks allow water
433 samples to be taken and the sources of nutrients in the groundwater body below CWs to be
434 identified. The local site hydrology (precipitation, groundwater table fluctuations and
435 evapotranspiration) has greater impacts on the pollutant removal by physical attenuation and by

436 biochemical transformations. Hydrogeochemical studies on an accurate spatial and temporal
437 resolution should explain the effects of precipitation on nutrient removal by dilution as well in
438 situ nutrient turnover. Effective CW management requires an understanding of the effects of
439 wetland hydrology on the physical and biochemical attenuation of nutrients in order to assess
440 their impacts on the surface emissions and subsurface export of nutrients and GHG. Data on the
441 species of N in groundwater below the CWs are required to provide an in-depth understanding of
442 wetland ecosystem services, particularly if CW systems have the potential to leak pollutants
443 down into the groundwater (Dzakpasu et al., 2014). Higher NH_4^+ concentrations in groundwater
444 below the CWs than the effluents are often reported (Harrington et al., 2007; Dzakpasu et al.,
445 2012). Therefore, questions arise with respect to NH_4^+ concentrations in groundwater below the
446 CWs if they have been transported from CWs. Linking geochemistry of groundwater below CWs
447 to site hydrology, water table fluctuations and soil/subsoil physico-chemical properties is required
448 to elucidate the major environmental drivers of C and N removal and or pollution swapping. The
449 quality of groundwater underlying CWs with regards to the Nr species is largely unknown.

450

451 7. Methodological Developments

452 Quantification of the occurrence and magnitude of N transformations and hydrochemical
453 properties is crucial to improve the assessment of CW ecosystem services and to minimize their
454 potential for pollution swapping. Measurement of GHG using the closed chamber method is
455 widely used but has large uncertainty in estimating the diurnal variability due to internal changes
456 in temperature and physical access to the chambers over 24 h time period. Gas ebullition and
457 diffusion measurements are quite challenging in CWs covered by vegetation because of the
458 difficulties in estimation of gas transfer velocity. Application of eddy covariance method is not
459 likely for most of the CWs as it requires larger surface area (> several ha) to avoid contribution of
460 surrounding area and complication of GHG foot printing. In addition, heterogeneity in CWs
461 surface increases uncertainty in better mixing of gas turbulence before it goes to the sensor.
462 Combination of chamber, ebullition and diffusion methods in a single system could minimise the
463 uncertainty in GHG estimation. Methane ebullition measurement was found to be similar to
464 surface emissions by chamber method but N_2O and CO_2 ebullition measurements were lower
465 than the surface emissions (Søvik et al., 2006). Use of *in situ* microcosm study and soil core
466 incubation methods can give better estimation of N_2O , CO_2 and CH_4 production and consumption
467 in light with the better GHG and C and N budgets. With the recent advancement of isotope
468 pairing and dilution techniques, single or simultaneously occurring C and N transformation
469 processes can be quantified in laboratory or *in situ* conditions (Huygens et al., 2013; Müller et al.,

470 2014). The isotope technique relies on the introduction of a known amount of ^{14}C and or ^{15}N into
471 the CW system and then quantification of C and N concentrations and isotopic compositions
472 through different C and N pools after incubation for a specific period. Laboratory methods
473 involve collection of intact soil/sediment cores, with subsequent incubation in the laboratory. The
474 *in situ* field techniques involve release of $^{14}\text{C}/^{15}\text{N}$ solution *in situ* in the CWs soils. Incubation of
475 intact soil cores with differentially labelled $^{15}\text{NH}_4^{14}\text{NO}_3$ and $^{14}\text{NH}_4^{15}\text{NO}_3$ can be used to quantify
476 the rates of different N transformation processes (Rütting and Müller, 2008). The quantification
477 of simultaneously occurring N transformation rates rely on the analysis with appropriate ^{15}N
478 tracing models. Development in the recent years in ^{15}N tracing techniques is now available,
479 which are able to identify process specific NO_2^- pools (Rütting and Müller, 2008), pathways
480 specific N_2O production and emission as well as $\text{N}_2\text{O}/\text{N}_2$ ratios (Müller et al., 2014). Traditional
481 techniques for investigation of gross N dynamics in sediments (Blackburn, 1979) could be
482 combined with the latest ^{15}N tracing techniques where all N transformation rates are included,
483 that are important in wetlands and under anoxic condition (Huygens et al., 2013). Thus, current
484 models should consider processes such as anammox and/or deammox and then be tested in CW
485 environments under various conditions. Denitrification in porewater samples can be measured by
486 analysing samples for dissolved N_2 in a membrane inlet mass spectrometer (MIMS; Kana et al.,
487 1994) and N_2O in gas chromatograph (GC; Jahangir et al., 2012). The studies of natural
488 abundance of ^{15}N and ^{18}O ($\delta^{15}\text{N}$ and $\delta^{18}\text{O}$) in NO_3^- is an insightful tool for the investigation of the
489 sources, fate and transformational processes of N in a system (e.g. in shallow groundwater, Baily
490 et al., 2011). The *in situ* NO_3^- push-pull method has been used to determine denitrification in
491 shallow groundwater (<3 m) in riparian wetlands (Addy et al., 2002; Kellogg et al., 2005) and in
492 deep groundwater in arable/grassland (Jahangir et al., 2013).

493
494 The isotope based techniques can also be extended to other elements e.g., a ^{33}P tracing model has
495 been developed recently to study phosphorous cycle in soil (Müller and Bünemann, 2014). These
496 techniques can be applied in the study of C, N and P biogeochemistry in aquatic environment. In
497 addition measurements of DOC and gases (CO_2 and CH_4) will provide insights into the C
498 consumption and transformation associated with the N transformations. Carbon and N dynamics
499 are influenced by the interacting effects of soil conditions with microbial community structure
500 and functioning. Microbial functioning involves transcription of genes, translation of messenger
501 RNA and activity of enzymes (Firestone et al., 2012). As such, activities of microbial
502 communities under various environmental conditions and how these contribute to C and N
503 dynamics is a very important area of future research (Müller and Clough, 2014). Molecular

504 approaches can be important tools to identifying and quantifying the genes that code for enzymes
505 mediating C and N cycles (Peterson et al., 2012). These tools help assess the relationships among
506 genes, environmental controllers and the rates of C and N processes. The scientific tools and
507 multidisciplinary techniques are now available to better understand C and N transformation rates,
508 processes and factors controlling the unwanted emission of N and C products to the environment.
509

510 8. Conclusions and Recommendations

511 The transformational processes on a mixture of contaminants within and below CWs can cause
512 pollution swapping. A holistic assessment of C and N dynamics in CWs is needed to fully
513 understand their removal, transport and impact on water quality and emissions to atmosphere.
514 Mixed contaminants entering CWs and those formed within and underneath CWs during
515 transformational processes must be considered in future studies. The overall balance of these
516 constituents will decide on whether a CW is a pollution source or a sink. This will necessitate a
517 higher degree of multi-level spatial and temporal monitoring and use of multi-disciplinary
518 techniques both *in* and *ex situ* to fully characterise all pathways of C and N loss. We cannot yet
519 suggest any best engineering aspect of CW type with the design optima in terms of nutrient
520 removal and GHG mitigation because the research based information are not yet abundant. For
521 this, transformation kinetics of C and N and net GHG emissions through all possible pathways
522 are required to provide a holistic assessment. However, combination of various types of CW
523 (several parallel VSSF CWs followed by HSSF CWs) and a mixture of plant types could provide
524 higher removal efficiency and lower GHG emissions. A conceptual model highlighting the state
525 of knowledge in this area and the specific research need to be done was presented in Figure 1.
526

527 Subsurface export of nutrients and GHG to groundwater through leaching through
528 convective/dispersion transport or by preferential flow paths created by dead roots and holes of
529 burrowing invertebrates and subsequent transport to surface water bodies should be accounted in
530 **CW management**. Reducing the saturated hydraulic conductivity below the wetland bed will help
531 reduce nutrients leaching to groundwater below the CWs cells. The reactive versus the benign
532 forms of the N transformation products should be evaluated. Data on **when, where and at what**
533 **rates** denitrification, deamox and anammox occur in CWs are needed as well as what are the key
534 **factors** that control such processes. The provenance of NH_4^+ in groundwater below CW cells and
535 its impact on down gradient receptors needs further elucidation. This review shows that CWs
536 have the potential to produce N_2O , DON, DOC, dissolved inorganic C (DIC), CO_2 and CH_4 and

537 that the GHG produced in CWs can be emitted to atmosphere. They can also be exported to fresh
538 waters via groundwater and degassed upon discharge to surface waters. Moreover, the DOC and
539 DIC transferred to the fresh water sediments (rivers and lakes) can produce GHG that in turn emit
540 to atmosphere. The amount of C and N exported from terrestrial ecosystem via the subsurface
541 pathway to fresh waters has been the missing piece of our understanding of global C and N
542 budgets. It is clear that data on the various C and N species along with the GHG in various CWs
543 systems are thus crucial to make a robust input-output balance of C and N in such a rising and
544 engineered ecosystem. Spatial and temporal variation of GHG emissions in CWs under different
545 management systems is critical to get much more rigorous estimates of emission factors. These
546 data will reduce the existing uncertainties in global C and N budgets.

547

548 Managing wetting and drying spells (pulsing hydrology) in CWs can enhance NH_4^+ removal.
549 Similarly, oxidation of organic C will increase CO_2 production and in anaerobic condition it can
550 be reduced to CH_4 . This requires more research into the C and N cycle processes over the wetting
551 drying spells which is now possible with the advancement in $^{14}\text{C}/^{15}\text{N}$ tracing and modelling
552 techniques. The selection of plant species is important to increase nutrients removal, sequester
553 more C and decrease GHG emissions which are an area that still requires more research across
554 types of CWs and countries. Further research is also still needed to be done on the impacts of
555 hydraulic retention time on nutrients dynamics and removal. Rates of nutrient accumulation or
556 fixation in soils and their *in situ* transformation in CWs need to be quantified to know their
557 contribution to C sequestration and GHG emissions.

558

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562

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564 The first author, M.M.R. Jahangir has reviewed articles in the relevant area, analysed results and
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567

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Table 1 Mineral N (mg N L^{-1}), total nitrogen (TN, mg N L^{-1}) and TN removal (%) in various CWs treating wastewater; average (\pm standard error, SE) are presented for TN removal; n.a – data not available

CW type	Treatment	N input (mg N L^{-1})			N output (mg N L^{-1})			N removal (%)			References
		TN	NH_4^+	NO_3^-	TN	NH_4^+	NO_3^-	TN	NH_4^+	NO_3^-	
SF_Finland	Municipal	1.4 ± 150	0.03 ± 5.8	0.3 ± 95	1.1 ± 48	0.01 ± 3.0	0.02 ± 6.7	21.4	66.7	93.3	Søvik et al., 2006
SF_Finland	Agril. runoff	66.1 ± 1.9	63.5 ± 1.3	0.7 ± 0.13	64.7 ± 1.7	61.2 ± 1.7	0.3 ± 0.09	2.1	3.6	57.1	Søvik et al., 2006
SF_Norway	Municipal	43.4 ± 3.6	41.5 ± 3.0	0.0 ± 0.0	36.7 ± 2.7	32.6 ± 1.9	0.9 ± 0.4	15.4	21.4	-800	Søvik et al., 2006
SF	Municipal	n.a	4.5	15.5	n.a	n.a	n.a	61	n.a	n.a	Song et al., 2011
SF	Domestic	n.a	40	5	n.a	n.a	n.a	97-98	n.a	n.a	Dzakpasu et al., 2011
SF	Various	n.a	39	4.4	n.a	n.a	n.a	39-48	n.a	n.a	Vymazal, 2007
SF	Municipal	n.a	36		n.a	n.a	n.a	39	n.a	n.a	Vymazal, 2010
SF	Municipal	n.a	196	<2	n.a	n.a	n.a	35	n.a	n.a	Shamir et al., 2001
SF	various	n.a	80	<1	n.a	n.a	n.a	>90	n.a	n.a	Harrington et al., 2007
SF	Municipal	n.a	0.95	1.54	n.a	n.a	n.a	45	n.a	n.a	Toet et al., 2005
SF	Dairy washout	227	n.a	n.a	n.a	n.a	n.a	40	n.a	n.a	Van der Zaag et al., 2010
All SF								31.3 ± 6.3			
HSSF_Estonia	Municipal	96.5 ± 3.0	83.9 ± 2.7	0.2 ± 0.02	46.2 ± 1.5	36.2 ± 1.4	5.9 ± 0.65	52.1	56.9	-2850	Søvik et al., 2006
HSSF_Norway	Municipal	53.4 ± 4.3	38.4 ± 7.7	14.1 ± 7.5	45.0 ± 4.1	43.1 ± 4.7	1.0 ± 0.8	15.7	-12.2	92.9	Søvik et al., 2006
HSSF	Dairy washout	$306 \pm 101^*$	n.a	n.a	$177 \pm 58^*$	n.a	n.a	42.2	n.a	n.a	Van der Zaag et al., 2010
HSSF	Domestic	n.a	74.9	3.9	n.a	n.a	n.a	29	n.a	n.a	O'Lunaigh et al., 2010
HSSF	Domestic	87						46-48			Mander et al., 2008
HSSF	Dairy washout	227						28			Van der Zaag et al., 2010
HSSF	Milk parlour	112	22	n.a	24	11	n.a	78	50	n.a	Kato et al., 2006
HSSF	Agriculture	67	40	0.85	27	11	1.1	47	39	-29	Vymazal and Kröpfelova, 2009
HSSF	Industry	124	65	8.5	103	31	7.4	20	20	8	Vymazal and Kröpfelova, 2009
HSSF	Landfill	157	149	1.5	147	98	1.3	30	33	31	Vymazal and Kröpfelova, 2009
HSSF	Municipal	43	24	2	24	14	1.2	40	30	33	Vymazal and Kröpfelova, 2009
All HSSF								40.4 ± 4.4			
VSSF_Estonia	Municipal	50.9 ± 9.2	35.7 ± 6.2	1.1 ± 0.32	43.1 ± 7.6	31.7 ± 5.5	1.7 ± 0.84	15.3	11.2	-54.5	Søvik et al., 2006
VSSF_Norway	Municipal	52.6 ± 5.2	49.6 ± 4.0	0.0 ± 0.0	47.8 ± 6.9	21.4 ± 6.9	25.5 ± 1.3	9.1	56.9	-25400	Søvik et al., 2006
VSSF	Municipal	41.0 ± 0.5	n.a	n.a	20.7 ± 0.8	n.a	n.a	49.3 ± 1.8	n.a	n.a	Yan et al., 2012
VSSF	Municipal	46 ± 13	n.a	n.a	n.a	n.a	n.a	74 ± 3	n.a	n.a	Zhao et al., 2014
All VSSF								37.0 ± 10.9			

SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow; * $\text{mg N m}^{-2} \text{h}^{-1}$

Table 2 Total organic C (TOC) removal (%) in various CWs treating wastewater; average (\pm standard error, SE) are presented for TOC removal; n.a - data not available

CWs type	Treatment	C input (TOC; mg C L ⁻¹)	C outflow (TOC; mg C L ⁻¹)	TOC Removal (%)	References
SF_Finland	Municipal	13.0 \pm 0.3	14.0 \pm 0.5	-7.7	Søvik et al., 2006
SF_Finland	Agril runoff	25.0 \pm 3.4	20.0 \pm 3.4	20.0	Søvik et al., 2006
SF_Norway	Municipal	26.7 \pm 2.9	17.1 \pm 1.8	36.0	Søvik et al., 2006
SF	Dairy wash out	186†	136†	27	Van der Zaag et al., 2010
All SF				18.8 \pm 9.4	
HSSF	Domestic	150*	n.a	n.a	Garcia et al., 2007
HSSF	Dairy wash out	186†	107.9†	42	Van der Zaag et al., 2010
HSSF_Estonia	Municipal	62.8 \pm 16.6†	41.0 \pm 11.3 †	34.7	Søvik et al., 2006
HSSF_Norway	Municipal	40.5 \pm 11.3	15.0 \pm 2.4	63.0	Søvik et al., 2006
All HSSF				46.6 \pm 7.3	
VSSF_Estonia	Municipal	132.2 \pm 32.2†	62.8 \pm 16.6 †	52.5	Søvik et al., 2006
VSSF_Norway	Municipal	40.5 \pm 11.3	15.0 \pm 2.4	63.0	Søvik et al., 2006
VSSF	Municipal	106 \pm 35	74 \pm 21	26 \pm 4.6	Yan et al., 2012
VSSF	Municipal	249 \pm 49	n.a	83 \pm 1.0	Zhao et al., 2014
All VSSF				56.2 \pm 9.5	

SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow; †BOD; *mg m⁻² h⁻¹

Table 3 Nitrous oxide (N₂O) emissions (mg N m⁻² d⁻¹); N₂ emissions (mg N m⁻² d⁻¹) and N₂O emission factor (N₂O/TN input*100) in various type of CWs; mean (± SE) was presented for N₂O emission factor; n.a - data not available

CW type	Treatment	Denitrification		N ₂ O-N/TN (%)	N ₂ -N/TN (%)	References
		N ₂ O emissions (mg N m ⁻² d ⁻¹)	N ₂ emissions (mg N m ⁻² d ⁻¹)			
HSF	Agril. tile drainage	0.01-0.12	n.a	0.19-1.4	n.a	Xue et al., 1999
HSF	Treated municipal	2.0±3.3	n.a	0.02-0.27	n.a	Johansson et al., 2003
HSF	Agril. drainage	-0.2-1.9	n.a	-0.14-0.52	n.a	Wild et al., 2002
HSF	Dairy wash out	16.8±7.0	n.a	0.33±0.12	n.a	Van der Zaag et al., 2010
HSF_Finland	Municipal	0.01±0.01	n.a	1.6±1.3	n.a	Søvik et al., 2006
HSF_Finland	Agril. runoff	0.40±0.25	n.a	0.37±0.18	n.a	Søvik et al., 2006
HSF_Norway	Municipal	4.0±1.6	n.a	1.5±4.4	n.a	Søvik et al., 2006
All SF		2.78±1.72		0.61±0.21		
HSSF	Domestic	0.2-17.0	n.a	0.06-3.8	n.a	Mander et al., 2005
HSSF_Estonia	Municipal	7.1±1.2	n.a	0.05±0.31	n.a	Søvik et al., 2006
HSSF_Norway	Municipal	6.9±4.3	n.a	0.24±0.53	n.a	Søvik et al., 2006
HSSF	Domestic	1.3-1.4	160-170	0.37-0.60	15.2-22.7	Mander et al., 2008
HSSF	Domestic	0.003-0.001	0.01-5.42	n.a	n.a	Teiter and Mander, 2005
HSSF	Domestic	0.13	n.a	0.008	n.a	Fey et al., 1999
HSSF	Dairy wash out	9.5±1.5	n.a	0.18±0.12	n.a	Van der Zaag et al., 2010
HSSF	Domestic	0.17	n.a	0.23	n.a	Liu et al., 2009
VSSF	Domestic	0.17	n.a	0.01		Mander et al., 2011
All HSSF		4.23±1.87		0.62±0.38		
VSSF	Domestic	0.001-0.002	0.01-5.0	n.a	n.a	Teiter and Mander, 2005
VSSF	Domestic	4.6	150	0.45-0.50	n.a	Mander et al., 2008
VSSF	Domestic	11.0	n.a	0.29	n.a	Mander et al., 2005
VSSF	Domestic	1.44	n.a	0.03		Mander et al., 2011
VSSF	Domestic	0.005	n.a	0.09	n.a	Gui et al., 2007
VSSF	Domestic	0.003	n.a	0.04	n.a	Liu et al., 2009
VSSF_Estonia	Municipal	15±3.9	n.a	04.3±0.95	n.a	Søvik et al., 2006
VSSF_Norway	Municipal	960±40	n.a	1.4±0.72	n.a	Søvik et al., 2006
All VSSF		123.8±106		1.01±0.48		

SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow

Table 4 Carbon dioxide (CO₂, mg C m⁻² d⁻¹), CH₄ (mg C m⁻² d⁻¹) and CH₄ emission factor (CH₄-C/TOC input*100) in various type of CWs; mean (± SE) was presented for CH₄ emission factor; n.a - data not available

CWs type	Treatment	CO ₂ emissions (mg C m ⁻² d ⁻¹)	CH ₄ emissions (mg C m ⁻² d ⁻¹)	CH ₄ / TC (%)	References
SF	Municipal	n.a	5.4	n.a	Tai et al., 2002
SF	Domestic	0.19	n.a	26	Gui et al., 2007
SF	Domestic	1.13	n.a	16	Liu et al., 2009
SF	Agril. drainage	n.a	0.88	31	Wild et al., 2002
SF	Dairy wash out	4250±550	223±35	9.45	Van der Zaag et al., 2010
SF_Finland	Municipal	1200±420	29±6.4	19±4.3	Søvik et al., 2006
SF_Finland	Agril runoff	3200±560	350±180	11±5.5	Søvik et al., 2006
SF_Norway	Municipal	1400±250	72±28	4.8±2.2	Søvik et al., 2006
All SF		1675±703	113±58	16.8±3.8	
HSSF	Domestic	n.a	1.7-528	n.a	Mander et al., 2005a
HSSF	Domestic	2.54-5.83	0.03-0.40	n.a	Teiter and Mander, 2005
HSSF	Domestic	5.33	0.001	0.03	Garcia et al., 2007
HSSF	Domestic	n.a	0.03	4.3	Gui et al., 2007
HSSF	Domestic	n.a	0.29	4.0	Liu et al., 2009
HSSF	Dairy wash out	3475±375	118±9.0	4.4	Van der Zaag et al., 2010
HSSF	Domestic	0.6-1.7	1.4- 4.1	0.12-0.23	Søvik et al., 2006
HSSF	Domestic	600	0.48	0.02	Mander et al., 2011
HSSF_Estonia	Municipal	3800±210	340±240	n.a	Søvik et al., 2006
HSSF_Norway	Municipal	790±170	130±43	9.5±3.3	Søvik et al., 2006
All HSSF		1010±672	112±74	3.23±1.4	
VSSF	Domestic	5.83-12.13	0.60-5.70		Teiter and Mander, 2005
VSSF	Domestic	n.a	16.4	n.a	Mander et al., 2005
VSSF	Domestic	n.a	0.013	1.68	Gui et al., 2007
VSSF	Domestic	n.a	0.13	1.73	Liu et al., 2009
VSSF	Municipal	2662 ± 175	33.5 ± 3.2	n.a	Mander et al., 2008
VSSF	Domestic	1080	3.36	0.05	Mander et al., 2011
VSSF_Estonia	Municipal	8400±2100	110±35	n.a	Søvik et al., 2006
VSSF_Norway	Municipal	22000±5000	140±160	0.39±0.27	Søvik et al., 2006
All VSSF		6616 ± 3779	42.9 ± 23.7	1.27±0.31	

SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow

Table 5 Nitrous oxide (N₂O, mg N m⁻² d⁻¹), CO₂ and CH₄ emissions (mg C m⁻² d⁻¹) in various type of CWs under different plant types; n.a - data not available

CW type	Wastewater type	Plant type	N ₂ O (mg N m ⁻² d ⁻¹)	CH ₄ (mg C m ⁻² d ⁻¹)	CO ₂ (mg m ⁻² d ⁻¹)	Reference
HSF	Secondary treated municipal	No plant	3.79±2.64	163±209		Johansson et al., 2003; Johansson et al., 2004
		<i>Typha lotifolia</i>	2.64±4.09	109±185	n.a	
		<i>Phalaris arundinacea</i>	3.79±3.44	212±151	n.a	
		<i>Glyceria maxima</i>	0.76±1.01	112±178	n.a	
		<i>Lemna minor</i>	1.45±1.18	450±182	n.a	
		<i>Spirogyra sp.</i>	0.98±1.25	107±135	n.a	
HSF	Sewage Treatment water	No plant	-0.26±2.53	-4.76±61.8	4.32±0.73	Ström et al., 2007
		<i>Typha atifolia</i>	4.94±2.00	225±47.7	25.3±4.08	
		<i>Phragmites australis</i>	7.80±2.53	333±76.6	25.1±4.74	
		<i>Juncus effusus</i>	3.87±1.86	489±46.3	26.1±3.00	
HSSF	Domestic	No plant	0.04±0.02	87±6.3	80±6.3	Maltais-Landry et al., 2009
		<i>Phragmites</i>	0.06±0.03	50±7.5	200±35	
		<i>Typha</i>	0.03±0.01	28±3.0	235±32	
		<i>Phalaris</i>	0.01±0.01	45±6.0	195±31	
VSSF	Municipal	<i>Phragmites australis</i>	15±3.9	110±35	8400±2100	Søvik et al., 2006
VSSF	Municipal	<i>Phragmites australis</i>	264	384		Mander et al., 2005

SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow

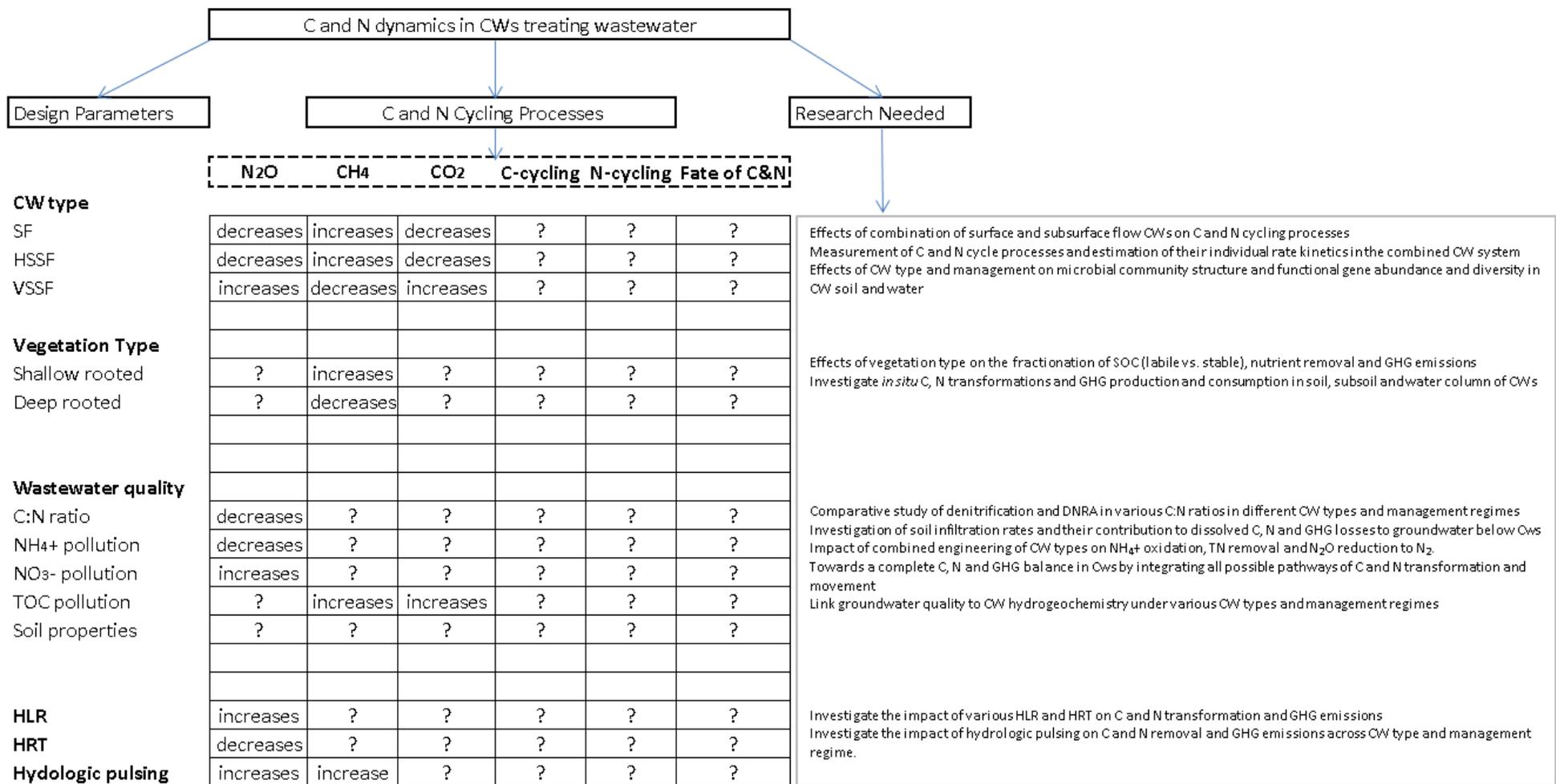


Figure 1 Conceptual model showing the current state of knowledge of C and N dynamics in constructed wetlands treating wastewater and the specific experimental work that needs to be undertaken in the future; SF- surface flow; HSSF – horizontal subsurface flow; VSSF- vertical subsurface flow; HLR – hydraulic loading rate; HTR – hydraulic retention time; ? – not known or very little known