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Interactive Comment

Interactive comment on "Carbon and nitrogen dynamics and greenhouse gases emissions in constructed wetlands: a review" *by* M. M. R. Jahangir et al.

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Response to the Referee's (Referee #2) Comments: MS. HESS-2014-272 The authors wish to thank Referee #2 for the very constructive comments and valuable suggestions. These have helped improve the manuscript:

Comments: Review for HESS Jahangir, Fenton, Gill, Müller, Johnston and Richards. Carbon and nitrogen dynamics and greenhouse gas emissions in constructed wetlands: a review. This manuscript sets out to review nitrogen and carbon cycling in constructed wetlands. I note however that the focus is much more strongly on nitrogen, rather than carbon (and therefore the title might benefit from a slight change?). Such a review would be very welcome and the manuscript could fill a clear gap in the literature. The manuscript is well-structured, detailed and in general a suitable style. There are however substantial corrections and amendments required (as I will detailed below) before it is publishable. So I suggest (and strongly encourage) major revisions and resubmit.

The corresponding author's detailed comments and corrections have been annotated in the pdf file, but in general: Comments 1. The manuscript is more of a literature summary, rather than a critical review. There are many places where data from a number of papers are summarized, listing a lot of numerical values. Sometimes this is done even when there is a table outlining the same data. I really look for critical analysis in reviews – this is currently missing. Responses: The manuscript has been revised thoroughly analysing the existing international literature and synthesising any knowledge gaps. Many of the numerical values in the text have been removed and any data repeated in tables were removed from the text. All the annotated comments and corrections of the referee have been incorporated into the text.

Comments 2. There are a number of times when research findings from a specific site and application, are generalized across all constructed wetlands. The authors should be very careful here. The manuscript reads (in quite a few places) that you have a specific site in mind and you are using the literature review to justify (often not particularly well) why you wish to use your site. But you are not stating what you site is, and this approach is of less relevance to readers. Responses: The specific system (site) based discussions and citations were removed to make it more generalised and to keep it more relevant to international readers. The idea for this review initially emerged from constructed wetlands treating agricultural and municipal waste waters. However the focus is now broader, considering that CWs are being used for the treatment of many other pollutants (landfilled leachate, Storm waters etc.). We now revised the manuscript incorporating a more generalised review of CWs.

Comments 3. There are many problems with sentence construction throughout the paper. Given that the majority of the authors have English as a first language, this is disappointing. Please do a serious edit for grammar and English. At times, your meaning is so hard to get to, as I had to pick apart sentences that did not really make sense. Responses: The manuscript has been edited thoroughly in this regard.

Comments 4. There is a methods section in the paper. While a (critical) review of methods is appropriate, I don't think the outline of an experimental setup is in any way appropriate for a review. Responses: We agree with the referee. The outline of the experimental set up has been removed.

Comments 5. There is a very generic statement after each section, saying something along the lines of "this area needs to be more fully investigated to get a better understanding of GHG emissions". This is too broad a statement. You have done a review of literature, so should be able to identify specific knowledge gaps and therefore research questions. This comes back to the lack of critical review. Responses: After analysing the existing literature, specific knowledge gaps have been identified and itemised for each section.

Comments 6. The final paragraph of the paper is very poor. In fact you conclude with an unfinished sentence!! Responses: The whole conclusion section including the final paragraph has been revised fully.

The specific comments of the referee #2 and responses to those comments (incorporated into the manuscript) have been provided below:

Comments: p7617 L 5: Be consistent with Nr or N(sub)r Responses: Nr has been used consistently throughout the text.

Comments: p. 7617, L 6: Not all CWs are sinks - as you show clearly in the rest of the manuscript. Careful about absolute statements. Responses: This statement has been rephrased. Now it reads 'There are natural sinks for Nr along the N cascade (Galloway

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et al, 2003; Tanner et al., 2005) but other sinks that are rehabilitated or artificial, may also be introduced and evaluated at key locations e.g. constructed wetlands (CWs) (Gold et al., 2013) or permeable reactive interceptors (Fenton et al., 2014).'

Comments: p. 7617 L21: add to be Responses: 'To be' has been added to the text

Comments: p. 7618, L 2-5: poor sentence structure Responses: The sentence has been rephrased 'Indirect N2O emissions from CWs effluents that discharge directly to aquatic environments have a default emission factor (EF) of 0.005 but with a wide range (0.0005-0.25). The wide range of EF is highlighting the large uncertainty, which warrants further refinement.'

Comments: p. 7618 L12-14: poor sentence structure Responses: The sentences have been rephrased and now reads 'For example, questions arise with respect to NH4+ concentrations in groundwater underlying the CWs (often higher in groundwater than the effluent e.g. Harrington et al., 2007; Dzakpasu et al., 2012).'

Comments: p. 7618 L15: components of what? Wetland or N cycle? Responses: The sentence has been re-written. It now reads 'Mass balance analysis of the different components of the N cycle and kinetics of the N transformation processes occurring within the treatment cells using the isotope-tracing 15N technique can provide mechanistic information for N transformation products (Lee et al., 2009; O'Luanaigh et al., 2010) and therefore can be used to start to answer such questions.'

Comments: p. 7618 L18-22: poor sentence structure Responses: The sentence has been re-written and now reads 'Huygens, et al. (2013) described two complementary stable isotope methods that can be used to determine N cycle processes in CWs: i) the isotope pairing and revised isotope pairing techniques which calculate total N2 production by denitrification and anaerobic ammonium oxidation (anammox) and ii) isotope dilution and tracing techniques which assess gross N transformation in wetland soils.'

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Comments: p. 7619 L4-6: too broad a statement Responses: This statement has been changed to make a more specific statement- ‘These methods in combination will be able to provide a comparative analysis of the rates of N transformation processes occurring in CWs. For example, in situ quantification of the rates of NH_4^+ oxidation and denitrification in CWs will give insights into their role in delivering NO_3^- and or NH_4^+ to groundwater and N_2O to the atmosphere.’

Comments: p. 7619 L13-15.: not all wetlands have peat. Responses: The word peat has been removed to keep the statement more generalised for wetlands. The sentence now reads ‘CO_2 is lost by plant respiration and aerobic organic matter decomposition, whilst CH_4 is lost by methanogenic activity during anaerobic decomposition of DOC.’

Comments: p. 7619 L 16: Not all wetlands are anoxic. And the ones that are anoxic, do not always release methane! Responses: The sentence has been revised and now reads ‘Methane is emitted from wetlands due to anoxic or suboxic conditions that occur in the soils.’

Comments: p. 7619 L : which N species? Responses: Here the N species is N_2O. The sentence has been rephrased and now reads ‘For example, van der Zaag et al. (2010) measured CH_4-C emissions as 0.2- 27% of the total C removed and N_2O emissions as 0.1- 1.16% of the total N removed in CWs.’

Comments: P 7620 L 22-28: This is just repeating material already shown in Table 1. Responses: This section was removed as suggested by the referee #1.

Comments: P 7621 L 10-19: Dont just repeat data already in a table. Responses: The date given in the table was removed from the text.

Comments: P7621 L 24-26: Again refrain from repeating text that is summarised in a table. Responses: These data were presented in the text in a more concise form and removed from the Table due to lack of sufficient information relevant to the removal efficiency. It now reads-‘In a single house domestic wastewater treatment using CWs, Gill et al. (2012) has shown mean removal efficiencies of 33-36% of total N (TN) but did not investigate the N cycling processes. To enhance such a low removal, the nature of N transformations and the fate of the removed N within the CWs should be investigated.'

Comments: P7622 L 5-7: Poor sentence structure. Responses: These sentences have been rephrased and now read 'However, contrasting results on the impacts of hydraulic loadings on nutrients removal efficiency are available in literature. Luo et al. (2005) reported that low HLR results in incomplete denitrification, whereas Zhang et al. (2006) argued that low HLR increases NH4+ and chemical oxygen demand (COD) removals by 25 and 11%, respectively.'

Comments: P7622 L 10-17: Synthesis required – don't just summarise data Responses: The sentences have been revised to synthesis the results and now read 'For example, Zhou and Hosomi (2008) constructed a surface flow wetland with a HLR of 0.20 m3 m−2 d−1 and an average water depth of 0.056 m. The authors estimated the N removal efficiency in terms of a comparison between N inputs and outputs but did not quantify how this rate corresponded to the different forms of the removed N. In addition, there was no examination if variations in HLR could increase the N removal capacity of the system whilst reducing pollution swapping by Nr. Similarly, Song et al. (2011) reported an average HRT of 2.4 days during wetland operation periods for the removal of NO3- and NH4+ without documented design optima or pollution swapping. Evaluation of systems in a holistic manner, including pollution swapping at different HRTs is important, particularly within the context of the changing hydrological cycle in a changing climate.'

Comments: P7622 L18: deduced how? Correct usage of "deduction"? Responses: The sentence has been rephrased and now reads 'Thus far, estimates of nutrient removal efficiencies are based on the subtraction of the concentrations of nutrients in the effluent from the influent, but the transformation kinetics of the removed nutrients are unknown.'

Comments: just a repeat of preceding sentence Responses: The repeated sentence has been removed from the text.

Comments: P7622 L 26: where does this come from? Suddenly you are talking about "pulsing"? Responses: All sentences have been restructured for better readability. These sentences now read-'The removal efficiency of pollutants in CWs depends on hydraulic loading rates (HLR) and hydraulic retention time (HRT) (Toet et al., 2005). The effects of HLR and HRT can vary with the nature of the use of CWs e.g. whether they are used for treating single or mixed contaminants. Therefore, to have the least amount of Nr delivery to the receiving waters or to the atmosphere, CWs need to be optimally designed with respect to HLR and HRT. Investigation into the effects of fluctuating hydraulic loadings (hydraulic pulsing) on N removal efficiency and its transformation products will provide information about the potential of pollution swapping for NO3-, NO2-, NH3, NH4+ and N2O.'

Comments: P7623 L 4: This has already been stated on p 7618, L 9 Responses: The sentence has been removed.

Comments: P23 L9-17: And again, we don't need a summary of data from previous papers - I'm looking for a synthesis and interpretation. Most of this paragraph suffers the same issue. Responses: The whole paragraph has been re-written analysing the existing knowledge and synthesising the knowledge gaps on nutrient accumulation and their fate in CWs. The paragraph now reads- 'The soil in CWs is a major sink for N. Data on the N accumulation (dissolved organic N- DON, TN, NH4+ or NO3–N) within the soil profile of various CWs are scarce as most studies focus on N balances between influent and effluent N loads. The accumulation of N could be a substantial part of the total N added to CWs; 30-40% (Shamir et al., 2001), 39% (Harrington et al., 2007), 9% (Mander et al., 2008) and 2.5% (Obarska-Pempkowiak and Gajewska, 2003). The wider range of N accumulation could be due to variations in CW types and their management. The accumulated species of N are rather reactive unless they are transformed to N2 by biogeochemical processes. For example, Shamir et al. (2001)

measured about 80% of the total accumulated N as organic and the remainder as NH_4^+. The organic N could be mineralised to NH_4^+ and NO_3^-, reactive forms of N, depending on the physico-chemical properties of soil. The Nr could be assimilated by plants and microbes living in CWs, which are recycled in a soil-plant-soil continuum. Typically, N accumulation has been found to decrease with soil depth: for example, Shamir et al. (2001) reported mean NH_4^+ concentrations in 0-0.15, 0.15-0.30 and 0.30-0.60 cm depths of 156, 151, and 28 mg N kg-1, respectively. However, dissolved nutrients can be preferentially leached down into deeper soil layers via different pathways e.g. root channels. In terms of the input-output balance, these are considered as removed N, but they remain in such a biogeochemically active system. In addition to N, organic C accumulation occurs in CW soils. In a gravel-bed CW, Nguyen (2000) measured 17.5, 16.2 and 3.6% of total C (TC) accumulation at surface, 0-10 and 10-40 cm depth. As such, CWs represent organic C and Nr rich systems where the products of the continuously occurring biogeochemical processes can be transported to fresh waters and to the atmosphere. The NO_3^- could be denitrified to N_2O or N_2 and NH_4^+ could be mineralised to NO_3^- or fixed in soil matrix. Estimation of the reaction kinetics of these processes is required to accurately measure the fate of the added nutrients in CWs. With methodological advancement, measurement of these processes is now possible. For example, application of 15N tracer technique ($^{15}NO_3^-$) can give insights into the NH_4^+ oxidation and fixation capacity of sediments below CWs. Estimating the rates of nutrient accumulation in soil and subsoils and their in situ transformation kinetics in various types of CWs is important. Elucidating the fate of these added nutrients will help to reduce their potential for pollution swapping which has been ignored so far in managing CWs. The stability of the accumulated C and N under changing climatic scenarios also needs to be addressed to consider the long term sustainability of CWs.'

Comments: P 24 L4: This review is reading more and more as if it were a literature review done to justify a specific PhD thesis topic. It seems to have the form: 1. Summarise existing literature with little synthesis/analysis. 2. Use the summary to justify an approach that was assumed at the beginning - the approach does not seem to flow from a literature synthesis. Responses: This section has been changed enormously to analyse the existing knowledge and to focus the knowledge gaps that are really very important. After a critical analysis, synthesis of new research questions was made. The whole section is appended below: 'Processes involved in N removal and N transformations in wetlands include sedimentation of particulates (Koskiaho, 2003); nitrification, denitrification and DNRA (Poach et al., 2003; Burgin et al., 2013), microbial assimilation and plant uptake-release (Findlay et al., 2003), anammox and DEAMOX (DEnitrifying AMmonium OXidation). Müller et al. (2014) developed a 15N tracing model, which is able to identify four different pathways of NO2- reduction to N2O: i) reduction of NO2- associated with nitriïňĄcation, ii) reduction of NO2- associated with denitriïňĄcation, iii) reduction of NO2- associated with organic N oxidation, and iv) co-denitriïňĄcation, a hybrid reaction where one N atom in NO2- originates from organic N and the other from NO2- reduction via denitriïňĄcation. Most of these pathways transfer Nr (mainly NH4+ and N2O) to the environment, however, some pathways can convert Nr to N2 (e.g. denitrification, anammox and DEAMOX). During denitrification, NO3- is used as a terminal electron acceptor to produce N2 or N2O (Starr and Gillham, 1993). Anammox can remove NO2- and NH4+ as N2 in CWs when the existing environment is hypoxic. The DEAMOX can remove NO3- and NH4+ as N2 where NO3- is converted to NO2- by autotrophic denitrification with sulphide (Kalyuznyi et al., 2006). Denitrification has been estimated to be a significant N removal process but actual quantification data are scarce. Limited studies have estimated N losses by denitrification e.g. 19% (Mander et al., 2008) and 86% (Obarska-Pempkowiak and Gajewska, 2003) of the total N input based on the mass balance study. To our knowledge, no data are available on denitrification measurements in soil/subsoils of surface flow CWs.

In addition, the two other processes that can remove Nr from the CWs (anammox and DEAMOX) are not well understood and so it is crucial to identify which of the processes are occurring in a specific type of CW and at what rate they occur. Once a process that provides N2 as the end product is determined in a specific system then the CW management could be directed towards enhancement of that process.

Hence, quantifying the rates of these processes under various CW types is required for improved N management towards lowering Nr in the environment. There is large uncertainty about the processes involved and their magnitude. Many studies focus on the mass balance approach and use the difference as the rate of N attenuation. N2O emissions from CWs have been reported to be high (EF 0.005; IPCC, 2014) but there is large uncertainty around these values due to the limited scope of the research that has been carried out to quantify such processes. The enhanced reduction of N2O to N2 needs further elucidation.

Similarly, C transformations involve respiration, fermentation, methanogenesis, CH4 oxidation and reduction of S, Fe and NO3-. Anerobic methane oxidation coupled with denitrification, a recently proposed pathway of the C cycle (á NorÃři and Thamdrup, 2014; Haroon et al., 2013; Islas-Lima et al., 2004), can reduce CH4 emissions in CWs. The C removal processes are sedimentation, microbial assimilation, gaseous emissions, dissolved C losses through water to ground and surface water bodies and chemical fixation (bonding with chemical ions). As CWs are designed to remove pollutants in an anaerobic/suboxic environment, they change the C and N dynamics and contribute significantly to CH4 and N2O emissions (Johansson et al., 2002, 2003; Mander et al., 2005, 2008; Stadmark and Leonardson, 2005; Liikanen et al., 2006). The hydrological, chemical and microbial processes of CWs are likely to be different from the processes occurring in more natural wetlands, because they receive nutrient rich waters from various sources. Increased nutrients and organics inputs will increase the productivity of wetland ecosystems and increase the production of GHGs. Søvic et al. (2006) measured N2O, CH4 and CO2 emissions in various CWs in different European countries and suggested that the potential atmospheric impacts of CWs should be examined as their development is increasing globally. Ström et al. (2007) recommended that CW management processes must consider the negative climatic aspects of increased emissions of GHGs in addition to their primary functions. Therefore, estimation of the contribution of CWs to global warming is required. Modelling or up-scaling of GHG emissions at national and regional scales to a global scale is important to improve global GHG budgets. The use of CWs to improve water quality is likely to increase to meet policy drivers such as the European Union Water Framework Directive and so the uncertainty of GHG budgets needs to be reduced to prevent pollution swapping.

In this regard, measurement of spatial and temporal variations (seasonal and diurnal) of GHG emissions is required. Moreover, plant mediated GHG emissions could be an important component of total emissions but research in this area is very limited. The GHG from CWs can vary with vegetated and non-vegetated systems. Vegetation and its composition affect the nutrient dynamics and the production, consumption and transport of greenhouse gases and hence their exchange between wetlands and atmosphere (Ström et al., 2003, 2005; Søvic et al., 2006; Johansson et al., 2003). Emergent plants can transport atmospheric O2 to rooting zone and contribute to C and N dynamics in wetland soils e.g. N2O and CO2 production and CH4 consumption (Brix, 1997). Vascular plants can exchange GHGs between the rooting zone and atmosphere (Yavitt and Knapp, 1998). Emissions of N2O and CH4 in CWs can vary across CW typologies e.g. surface flow or subsurface flow (Van der Zaag et al., 2010). Therefore, the assessment of GHG emissions in various types of CW (surface flow, subsurface flow; vertical and horizontal), under different management systems (vegetated, nonvegetated, plant species composition) and usage (municipal waste water, agricultural runoff, landfill leachate) is necessary in light of the national and global GHG budgets and mitigation of GHG emissions. In addition, such measurements will help scientists, environmental managers and policy makers to adopt environmental friendly construction and management of CWs. Assessment of the reactive versus the benign forms of C and N transformation products in various types of CWs will give insights into their environmental friendly design and management.'

Comments: P7626 L 14: what do you mean by drainage, and by fluxes, and by "drainage fluxes" Do you mean "export"? Careful about the language you use please. Responses: The words drainage fluxes were replaced with export.

Comments: P7626 L26-27: so re there key methodological research questions that result from your lit review? You have not really made a case for methodological constraints. Which ecosystem? Do you have one in mind? Responses: In fact, there is no methodological constraint to conduct research on C and N dynamics in groundwater below the CWs. However, the whole section was re-written to make it more precise and clear. We don't have any specific site in mind but here 'this' ecosystem means the constructed wetland ecosystem. Now it is corrected in the text. The section now reads- 'Dissolved GHGs in porewater in wetland soils and subsoils can be emitted to the atmosphere by transpiration of vascular plants (from within the rooting zone) and via groundwater, upon discharge to the surface waters. Dissolved GHGs in groundwater can flow towards surface waters by advective transport and or by dispersion of groundwater. The GHGs produced in soils/subsoils in CWs can also be emitted to the atmosphere by ebullition and diffusion. Elberling et al. (2011) reported that in wetlands, the transport of subsurface soil gases occurs both via diffusive transport in the pores and through the vascular plants. In addition to measuring the surface emissions, Ström et al. (2007) also measured a considerable quantity of CH_4 in porewater and found a significant correlation ($p = 0.021$) between the surface emissions and porewater CH_4 concentrations in vegetated wetlands. Measuring porewater GHGs and linking these to the surface emissions and subsurface export to groundwater below CWs will help to estimate a better GHG balance from both a national and global context. Elberling et al. (2011) linked subsurface gas concentrations in wetlands to the surface fluxes using a diffusion model which has demonstrated the need for future studies on subsurface GHG production, consumption and net GHG emissions in CWs ecosystem in a climate change context. It is important to characterise soils and subsoils physical and hydraulic properties and to assess their potential to percolate dissolved nutrients and gases to the underlying groundwater. To our knowledge, this indirect pathway of GHG emissions from CWs has never been reported despite the fact that this would appear to have a high biogeochemical potential to produce and exchange GHGs. The balance between N and C input and output flows between CWs and aquatic and atmospheric environments together with the direct and indirect emissions of C and N species could be an important input to global C and N budgets.'

Comments: P7627 L9-12: Review this paragraph for English and sentence structure. Responses: The paragraph has been amended as follows: 'CWs can be designed with or without a clay liner or a compacted soil bed at the base, which can lead to large differences in permeability of the underlying layers. The variation in permeability of a CW soil bed will affect solute, nutrient and GHG flows and their interactions with the underlying groundwater (Dzakpasu et al., 2012; 2014). Groundwater hydrogeochemistry below CWs can therefore provide a unique insight into such interactions. An example of such interactions would be between nutrient rich water discharging from CW cells mixing with laterally moving regional groundwater. It should be noted that groundwater can also discharge into CWs depending on the hydraulic gradients. This necessitates that multiple multi-level piezometers or boreholes are installed at such sites to elucidate groundwater flow direction, hydraulic gradients and hydraulic permeability. Such monitoring networks allow water samples to be taken and then sources of nutrients in the groundwater body below CWs can be identified. For example, natural abundance of N (δ15N) and oxygen (δ18O) can be used to identify the sources of NO3- and infer transformational processes responsible for concentrations in groundwater (Baily et al., 2011). The local site hydrology (precipitation, groundwater table fluctuations and evapotranspiration) has greater impacts on the pollutant removal by physical attenuation and by biochemical transformations. Hydrogeochemical studies on an accurate spatial and temporal resolution should explain the effects of precipitation on nutrient removal by dilution as well in situ nutrient turnover. Despite conditions within CWs being saturated throughout the year, changes in the water table elevation affect the oxygen (O2) concentrations in CWs which is key parameter for the biogeochemistry of soils and subsoils. For example, lowering the water level increases the O2 concentrations and accelerates soil OM decomposition, thereby increasing CH4 oxidation and CO2 emissions. Highly contrasting results on gas emissions with fluctuating water levels have been reported and the controlling mechanisms are unclear (Elberling et al., 2011). Effective CW management requires an understanding of the effects of wetland hydrology on the physical and biochemical attenuation of nutrients in order to assess their impacts on the surface emissions and subsurface export of nutrients and GHGs. Data on the species of N in groundwater below the CWs are required to provide an in-depth understanding of wetland ecosystem services, particularly if CW systems have the potential to leak pollutants down into the groundwater (Dzakpasu et al., 2014). A study on a clay lined CW system reported that the nutrient content in the underlying groundwater to be NH4+-N 4.0 mg L-1, NO3–N 0.2 mg L-1and molybdenum reactive phosphorus (MRP) <0.01mg L-1. The elevated NH4+-N indicates transport of nutrient from the CW to groundwater through leakage or in situ N transformations such as DNRA. However, the impact of CWs on the local underlying groundwater quality has been seldom assessed. Linking geochemistry of groundwater below CWs to site hydrology, water table fluctuations and soil/subsoils physico-chemical properties is required to elucidate the major environmental drivers of C and N removal and or pollution swapping.'

Comments: P7628 L10: But you've been through these processes in the previous section? Responses: This section has been deleted as these processes have already been discussed in previous sections. Referee #1 also has suggested the removal of this section.

Comments: P7635 L17-18: poor sentence structure Responses: The sentence has been re-structured and the whole paragraph has been revised. This paragraph now reads – 'Quantification of the occurrence and magnitude of N transformations and hydrochemical properties is crucial to improve the assessment of CW ecosystem services and to minimize their potential for pollution swapping. With the recent advancement of isotope pairing and dilution techniques, single or simultaneously occurring N transformation processes in CWs can be quantified in laboratory or in situ conditions (Huygens et al., 2013; Müller et al., 2014). The isotope technique relies on the introduction of a known amount of 15N into the CW system and then quantification of N concentrations and isotopic compositions through different N pools after incubation for a specific period. Laboratory methods involve collection of intact soil/sediment cores, with sub-

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sequent incubation in the laboratory. The in situ field techniques involve release of 15N solution in situ in the CWs soils. The in situ stable isotope techniques may be an appropriate tool for the determination of simultaneously occurring N transformation processes in wetland soils (Huygens et al, 2013).

Details of the application of isotope pairing/ revised isotope pairing techniques and isotope dilution/ tracing techniques have been presented elsewhere (Huygens et al., 2013). In brief, incubation of intact soil cores with differentially labelled 15NH414NO3 and 14NH415NO3 can be used to quantify the rates of different N transformation processes ((Rütting and Müller, 2008). The quantification of simultaneously occuring N transformation rates rely on the analysis with appropriate 15N tracing models. Development in the recent years in 15N tracing techniques is now available, which are able to identify process specific NO2- pools (Rütting and Müller, 2008), pathways specific N2O production and emission as well as N2O/N2 ratios (Müller et al., 2014). Traditional techniques for investigation of gross N dynamics in sediments (Blackburn, 1979) should be combined with the latest 15N tracing techniques where all N transformation rates are included that are important in wetlands and under anoxic condition (Huygens et al., 2013). Thus, current models should consider processes such as anammox and/or DEAMOX and then be tested in CW environments under various conditions. The in situ NO3- push-pull method has been used to determine denitrification in shallow groundwater (<3 m) in riparian wetlands (Addy et al., 2002; Kellogg et al., 2005) and in deep groundwater in arable/grassland (Jahangir et al., 2013). This method can be used in CW sites using piezometers or boreholes screened at different intervals to investigate in situ C and N dynamics. In this method the parameters of importance to be analysed are 15N2O, 15N2, 15NO3-, and 15NH4+. In addition measurements of DOC and gases (CO2 and CH4) will provide insights into the C consumption and transformation associated with the N transformations.

Carbon and N dynamics are influenced by the interacting effects of soil conditions with microbial community structure and functioning. Occurrence of C and N cycling processes are controlled by the environment which involves transcription of genes, translation of messenger RNA and activity of enzymes (Firestone et al., 2012). As such, activities of microbial communities under various environmental conditions and how these contribute to C and N dynamics is a very important area of future research (Müller and Clough, 2013). Molecular approaches can be important tools to identifying and quantifying the genes that code for enzymes mediating C and N cycles (Peterson et al., 2012). These tools help assess the relationships among genes, environmental controllers and the rates of C and N processes.'

Comments: P 7636 L6: What is this Methods section doing in a review article? Totally out of place. Responses: This method section has been removed.

Comments: P7638 L17-19: poor sentence structure Responses: The sentence has been rephrased and now reads 'The transformational processes on a mixture of contaminants within and below CWs can cause pollution swapping. A holistic assessment of C and N dynamics in CWs is needed to fully understand their removal, transport and impact on water quality and emissions to the atmosphere. Mixed contaminants entering CWs and those formed within and underneath CWs during transformational processes must be considered in future studies. The overall balance of these constituents will decide on whether a CW is a pollution source or a sink. This will necessitate a higher degree of multi-level spatial and temporal monitoring and use of multi-disciplinary techniques both in and ex situ to fully characterise all pathways of C and N loss.'

Comments: P7638 L24: gradient of what? Responses: gradient of geochemistry. The sentence is corrected and reads 'Leakages of nutrients and gases from CWs to groundwater can only be elucidated through the techniques itemised herein and a better understanding of the transformation processes along a vertical geochemical gradient from the CW into the underlying aquifer.'

Comments: P7639 L1: This is just a summary of previous text. Need more than this for Conclusions. Responses: This section has been revised to provide a better understanding of what has been discussed from the introduction through to the discussion and is now related to the objectives of the study. In now appears as: 'The reactive versus the benign forms of the N transformation products should be evaluated in various CWs. An understanding of how N removal occurs and how losses of N and associated gases impact water and air quality is required. Denitrification, DEAMOX and anammox are the processes which remove N to its benign form (N2) and all other processes produce only reactive forms of N. Data on when, where and at what rates denitrification, DEAMOX and anammox occur in CWs are needed as well as what are the key factors that control such processes. The provenance of NH4+ in groundwater below CW cells needs clarification and its impact on down gradient receptors.

This review shows that CWs have the potential to produce N2O, DON, DOC, DIC, CO2 and CH4 and that the GHGs produced in CWs can be emitted to the atmosphere. They can also be exported to fresh waters via groundwater and degassed upon discharge to surface waters. Moreover, the DOC and DIC transferred to the fresh water sediments (rivers and lakes) can produce GHGs that in turn get emitted to the atmosphere. Therefore, it is important to know the concentrations of dissolved C and N species and the rates of production and consumption of GHGs in groundwater below CWs. The amount of C and N exported from terrestrial ecosystem via the subsurface pathway to fresh waters has been the missing piece of our understanding of global C and N budgets. It is clear that data on the various C and N species along with the GHGs in various CWs systems are thus crucial to make a robust input-output balance of C and N in such a rising and engineered ecosystem. Spatial and temporal variation in N2O emissions in CWs under different management systems is critical to get much more rigorous estimates of N2O emission factors. These data will bring down the existing uncertainties in global C and N budgets.

Managing wetting and drying spells (pulsing hydrology) in CWs can enhance mineralisation of organic N and oxidation of NH4+ to NO2-/NO3- and then denitrification/anammox could transform these species to benign N2. This requires more research into the N cycle processes over the wetting drying spells which is now possible with the advancement in 15N tracing and modelling techniques. With the recent advancement of isotope pairing and dilution techniques single or simultaneously occurring N transformation processes can be quantified. The isotope based techniques can also be extended to other elements e.g., a 33P tracing model has been developed recently to study phosphorous cycle in soil (Müller and Bünemann, 2014). Further reducing the saturated hydraulic conductivity below the wetland bed will help reduce nutrients leaching to groundwater below the CWs cells. The selection of plant species is important to increase nutrients removal, sequester more C and decrease greenhouse gas emissions which is an area that still requires more research across types of CWs and countries. More research is also still needed to be done on the impacts of hydraulic retention time on nutrients dynamics and removal. Subsurface export of nutrients and GHGs to groundwater through leachates, preferential flow paths created by dead roots and holes of burrowing invertebrates and subsequent transport to surface water bodies should be accounted in CW management. Rates of nutrient accumulation or fixation in soils and their in situ transformation in CWs need to be quantified to know their contribution to C sequestration fate in the environment. Estimation of GHG production in CWs, their consumption and emissions to the atmosphere in a changing climate is urgent.'

Comments: P7640 L4: Unfinished last sentence!!! What a way to finish a submitted manuscript. This is a very poor concluding paragraph!!! Responses: The sentence was complete in the submitted manuscript but had been cut off during pdf construction for some reason. It now reads as follows: – 'The ecosystems services from CW will make them an attractive option for water pollution mitigation while providing an important aquatic habitat, but their integrated impact on environmental quality needs to be addressed in a holistic manner to avoid unwanted side effects.'

Please also note the supplement to this comment:
http://www.hydrol-earth-syst-sci-discuss.net/11/C4411/2014/hessd-11-C4411-2014-

HESSD

11, C4411–C4429, 2014

supplement.pdf

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 11, 7615, 2014.