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EMISSION RATES OF N2O, N2, CH4 AND CO2 IN RIPARIAN GREY ALDER FORESTS AND SUBSURFACE FLOW CONSTRUCTED WETLANDS

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CONTENTS

ORIGINAL PUBLICATIONS

This thesis is based on the following papers, which are included as appendices at the end of the thesis:

I   **Teiter, S.,** Augustin, J., Lõhmus, K. and Mander, Ü. 2006. Comparative study of N_2O, N_2 and CH_4 emission rates in alder stands in Estonia and Germany. In: Mander, Ü. and Lõhmus, K. (Eds.) Riparian alder forests: Their importance as buffer zones and bioenergy sources. Kluwer Academic Publishers, Dordrecht. (Accepted).

II   **Teiter, S.** and Mander, Ü. 2005. Emission of N_2O, N_2, CH_4 and CO_2 from constructed wetlands for wastewater treatment and from riparian buffer zones. Ecological Engineering 25(5), 529–542.

III Mander, Ü., Kuusemets, V., Lõhmus, K., Mauring, T., **Teiter, S.** and Augustin, J. 2003. Nitrous oxide, dinitrogen, and methane emission in a subsurface flow constructed wetland. Water Science and Technology 48(5), 135–142.

IV Mander, Ü., Lõhmus, K., **Teiter, S.,** Kuusemets, V., Mauring, T., Uri, V. and Augustin, J. 200X. Gaseous nitrogen emission and nitrogen removal efficiency in riparian alder forests and subsurface flow constructed wetlands. Ecology. (Submitted).

V   Mander Ü., Lõhmus, K., Kuusemets, V., Ivask, M., **Teiter, S.** and Augustin J. 2005. Budgets of nitrogen and phosphorus fluxes in riparian grey alder forest. In: Vymazal, J. (Ed.) Natural and Constructed Wetlands: Nutrients, Metals and Management. Backhuys Publishers, Leiden, 1–19.

VI Mander, Ü., Lõhmus, K., Kuusemets, V., **Teiter, S.** and Nurk, K. 200X. Dynamics of nitrogen and phosphorus budgets in a horizontal subsurface flow constructed wetland. Water Research. (Submitted).

Author's contribution

Publications I and II: The author is fully responsible for the fieldwork, data collection and analysis and fully responsible in writing the manuscript.

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Publications V and VI: The author is responsible for the fieldwork and data collection considering gas sampling and analyses.

ABSTRACT

Teiter, S. 2005. Emission rates of N_2O, N_2, CH_4 and CO_2 in riparian alder forests and subsurface flow constructed wetlands.

Constructed wetlands (CW) for wastewater treatment and riparian buffer zones are important ecotechnological measures to control water quality in agricultural catchments. At the same time, these ecosystems have been thought to be hot spots of greenhouse gas emissions. However, only a small amount of research has been conducted on studying this process. Therefore, nitrous oxide (N_2O), dinitrogen (N_2), methane (CH_4), and carbon dioxide (CO_2) fluxes in three study sites in Estonia: a riparian alder stand (Porijõgi) and in horizontal and vertical flow constructed wetlands (Kodijärve and Kõo) were measured using the "closed chamber" and He-O methods. The replicates' average values of N_2O, N_2, CH_4 and CO_2 fluxes from the riparian grey alder stand varied from -3.3 to 180 µg N_2O-N m^{-2} h^{-1}, 0.02 to 17.38 mg N_2-N m^{-2} h^{-1}, -5.9 to 805 µg CH_4-C m^{-2} h^{-1} and -3.9 to 290 mg CO_2-C m^{-2} h^{-1} respectively. Dinitrogen emission has been found to be the most important component in nitrogen retention from the studied riparian grey alder forest (up to 509 kg N_2-N ha^{-1} $year^{-1}$). Considering all inputs and outputs, N removal efficiency in grey alder stands seems to slow down with increasing stand/tree age. At the same time, the immobilization of N in soil is increasing. This suggests that grey alder buffer communities should be managed by regeneration cutting, which removes part of the nutrients, and helps to keep the N removal rate high.

In CWs the values of N_2O, N_2, CH_4 and CO_2 fluxes were 1 to 2600 µg N_2O-N m^{-2} h^{-1}, 0.2 to 130 mg N_2-N m^{-2} h^{-1}, 1.7 to 87200 µg CH_4-C m^{-2} h^{-1} and -6.1 to 1050 mg CO_2-C m^{-2} h^{-1} respectively. The release of all gases studied was significantly higher during the warmer period, although no significant correlation was found between N_2O flux and soil/water temperature. Similar to the purification performance, gaseous emissions in spring and early summer were significantly lower than in autumn. The most intensive flux of N_2O and CH_4 was observed in the chambers installed above the inflow pipes of horizontal flow beds. The vertical flow wetland did emit significantly more N_2O than the horizontal flow beds. On the other hand, N_2 is the largest component in N removal from CWs (up to 2500 kg N_2-N ha^{-1} $year^{-1}$). Water table level was found to be the most significant parameter of CWs regarding gas emissions, showing a significant positive Spearman rank order correlation with N_2O flux and a significant negative rank correlation with CH_4 emission.

The global warming potential (GWP) from N_2O and CH_4 was comparatively high in both types of CWs (4.8 ± 9.8 and 6.8 ± 16.2 t CO_2 eq ha^{-1} a^{-1} in the horizontal subsurface flow (HSSF) CW and 6.5 ± 13.0 and 5.3 ± 24.7 t CO_2 eq ha^{-1} a^{-1} in the hybrid CW respectively). The GWP of the riparian alder forest from both N_2O and CH_4 was relatively low (0.4 ± 1.0 and 0.1 ± 0.30 t

CO$_2$ eq ha^{-1} year^{-1} respectively), whereas the CO$_2$-C flux was remarkable (3.5 ± 3.7 t ha^{-1} year^{-1}). The global influence of CWs is not significant. Even if all global domestic wastewater were treated by wetlands, their share of the trace gas emission budget would be less than 1%.

1. INTRODUCTION

1.1. The role of riparian alder forests and subsurface flow constructed wetlands

Riparian buffer zones and constructed wetlands (CW) for wastewater treatment are important ecotechnological measures to control water quality in agricultural catchments (Peterjohn and Correll, 1984; Kadlec and Knight, 1996; Kuusemets and Mander, 1999). Denitrification, which is generally referred to as the microbial reduction of NO_3 -N to NO_2 -N and further to gaseous forms: NO, N_2O and N_2 (Knowles, 1982), has been found in numerous studies to be a significant process in nitrogen removal in riparian buffer zones (Groffman et al., 1991; Hanson et al., 1994; Weller et al., 1994; Gold et al., 1998; Hefting and de Klein, 1998; Groffman et al., 2000). In the majority of these studies, nitrous oxide (N_2O) fluxes have been measured, and only a few studies pay attention to dinitrogen (molecular nitrogen; N_2) emission (Blicher-Mathiesen et al., 1998; Watts and Seitzinger, 2000; Butterbach-Bahl et al., 2002). N_2O, as one of the greenhouse gases, is increasing in the atmosphere at a rate of about 0.3% $year^{-1}$ (Mosier, 1998). It has an atmospheric lifetime of about 120 years, a global warming potential of 296 relative to CO_2, over a 100-year time horizon, and is responsible for about 5% of the anticipated warming (IPCC, 2001). Riparian zones have the potential to be hotspots of N_2O production in the landscape (Groffman et al., 2000). The rate of N_2 and N_2O emission is necessary to measure regarding the global warming effects (Denmead and Raupach, 1993; Groffman et al., 2000; Watts and Seitzinger, 2000; Mander et al., 2003, 2005b; Teiter and Mander, 2005). Adequate knowledge of conditions influencing this rate can help to minimize N_2O emissions by landscape planning and management (Augustin et al., 1998b). Likewise, riparian wetlands and wet riparian forests can be sources of methane (CH_4; Jones and Mulholland, 1998; Rusch and Rennenberg, 1998), which is another greenhouse gas increasing in the atmosphere at a rate of about 0.8% $year^{-1}$ (Mosier, 1998). Methane in the atmosphere has a lifetime of 8.4 years. On a 100-year time horizon, CH_4 has a global warming potential of 23 relative to CO_2, and is responsible for about 20% of the anticipated warming (IPCC, 2001). Both denitrification and methane formation depend on the oxygen status of the soil or sediment. As a result, the spatial and temporal variability of fluxes of both N_2O (Robertson and Tiedje 1984; Ambus and Christensen 1993; Augustin et al., 1998b; Gold et al., 1998; Jacinthe et al., 1998) and CH_4 (Saarnio et al., 1997; Willison et al., 1998) are extremely high. Denitrification rates in soils are also influenced by carbon availability, nitrate availability, temperature, and pH (Mitsch and Gosselink 1993). CH_4 is produced in anoxic soils and sediments, while well-drained soils act as a sink for atmospheric CH_4 due to methane oxidation (negative emission), through either ammonia oxidizers or methanotrophs (Hanson et al., 1994).

Contrary to riparian buffer zones and natural wetlands, far fewer studies have been carried out concerning N_2O and CH_4 fluxes from CWs for wastewater treatment. Most of the available data concern the contribution of free water surface CWs to N_2O (Lund, 1999; Xue et al., 1999; Bachand and Horne, 2000; Lund et al., 2000; Spieles and Mitsch, 2000; Wild et al., 2002; Johansson et al., 2003) and CH_4 (Tanner et al., 1997; Tai et al., 2002; Wild et al., 2002) emissions. Only two works (Fey et al., 1999; Tanner et al., 2002) have considered the nitrous oxide fluxes from subsurface flow CWs. Dinitrogen emission from CWs has been measured by Mander et al. (2003, 2005b) and Teiter and Mander (2005).

Numerous studies consider emissions and sequestration of carbon dioxide (CO_2) in wetlands (Mitsch and Gosselink, 1993; Funk et al., 1994; Hamilton et al., 1995; Lafleur et al., 1997; Joiner et al., 1999; Griffis et al., 2000; Christensen et al., 2003). Depending on meteorological and hydrological conditions, wetlands can be sources or sinks of carbon (Clark et al., 1999; Waddington and Roulet, 2000; Whiting and Chanton, 2001; Arneth et al., 2002).

1.2. Objectives

The objectives of the current PhD dissertation are: (1) to quantify and compare N_2O, N_2, CH_4 and CO_2 emission rates in a grey alder stand (Porijõgi), and two CWs (Kodijärve and Kõo) for municipal wastewater treatment, using two different methods – the "closed chamber" method and the He-O method; (2) to determine the significance of factors controlling the N_2O, N_2, CH_4 and CO_2 fluxes from the grey alder stand and CWs; (3) to evaluate the global warming potential (GWP) of analysed greenhouse gases from riparian buffer zone and constructed wetlands; (4) to estimate the role of N_2O and N_2 emission in nitrogen budgets in the Porijõgi riparian grey alder stand and the Kodijärve HSSF filter CW.

2. MATERIALS AND METHODS

2.1. Description of studied alder stand

Experiments for measuring gas emissions were carried out in three grey alder riparian sites (Porijõgi WET, Porijõgi DRY and Porijõgi EDGE) in southeastern Estonia (Porijõgi; Fig. 1C).

The Porijõgi riparian buffer zone site is a grey alder stand situated in the moraine plain of southeast Estonia (Tartu County, Sirvaku (58º 13` N, 26° 47` E)) on the right bank of a small river, the Porijõgi, which flows in a primeval valley where agricultural activities ceased in 1992. The vegetation of the study site was described using the Braun-Blanquet cover scale (Kent & Coker, 1996). *Aegopodium podagraria* and *Impatiens noli-tangere* were predominant in Porijõgi *Alnus*-sites (Mander et al., 1995). The soil type in the Porijõgi grey alder stand was Mollic-Eutric Gleysol. The main soil characteristics of the Porijõgi grey alder stand are presented in Table 1.

Table 1. Soil characteristics of the riparian study sites (Teiter et al., accepted, Publication I)

Study site	Average water table	pH	Dry substance %	N %	NH_4^+ mg/100g	NO_3^- mg/100g	C %
Porijõgi WET	0–10 cm	6.5	55.0	0.4	1.88	0.09	4.0
Porijõgi DRY	45–95 cm	6.3	44.4	0.8	0.33	0.14	4.5
Porijõgi EDGE	45–95 cm	6.3	65.5	0.3	0.61	0.11	5.3

2.2. Description of studied constructed wetlands

According to alder stand, the gas emissions in two types of CWs were measured. These wetlands are located in the southern part of Estonia. The Kodijärve horizontal subsurface flow (HSSF) planted sand filter (constructed in October 1996) purifies the wastewater from a hospital for about 40 population equivalents. The system consists of two beds (chambers), each 25×6.25×1 m, which are filled with coarse sand. In May 1997, the right bed was planted with *Typha latifolia* and the left bed with *Iris pseudacorus* and *Phragmites australis*. During the following development, the *Typha* and *Iris* have almost disappeared, and the right bed is predominantly covered by *Scirpus sylvaticus*, while *P. australis* predominates in the left bed (Fig 1A). Due to the higher clay content in the filter material, the left bed (below also referred to as "wet bed") has somewhat wetter and less aerated conditions than the right bed ("dry bed"; Mander et al., 2001).
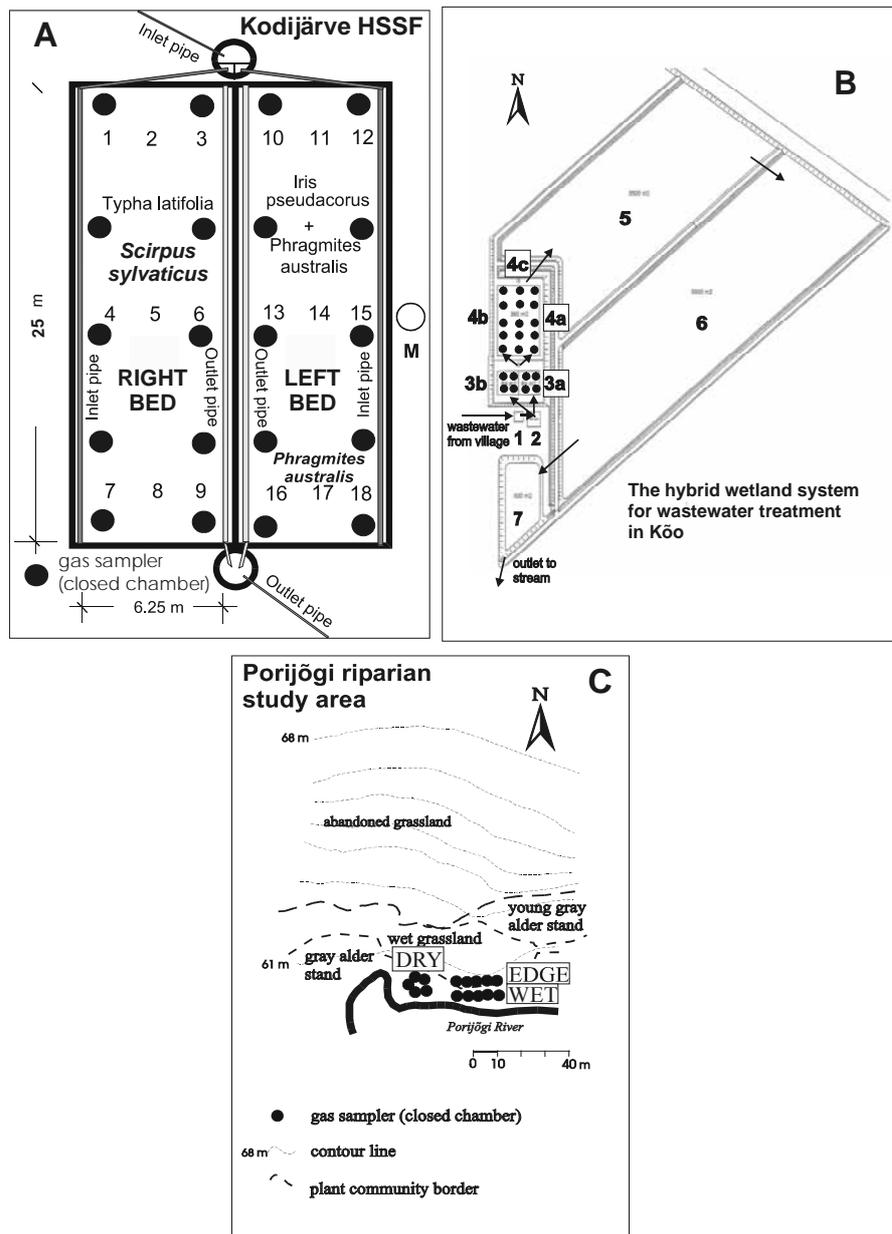
Figure 1. Diagrams of study sites: A – horizontal subsurface flow planted sand filter (HSSF) in Kodijärve, B – hybrid wetland system in Kõo, and C – riparian study area in Porijõgi. In part A: plant names in italics indicate the present dominant species in the beds, M - automatic weather station. In part B: 1 – pumping station; 2 – septic tank; 3 – vertical subsurface flow filter (VSSF; 2×64 m^2), a – right part, b – left part; 4 – HSSF (365 m^2), a – left inflow, b – right inflow, c – outflow; 5 – 1st free water surface wetland (FWSW; 3600 m^2), 6 – 2nd FWSW (5500 m^2), 7 – polishing pond (500 m^2).

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The hybrid treatment wetland system in Kõo consists of a two-bed vertical subsurface flow (VSSF; filter (2×64 m^2, filled with 5–10 mm crushed limestone, planted with *P. australis*), an HSSF filter (365 m^2, filled with 15–20 mm crushed limestone, planted with *T. latifolia* and *P. australis*), and two free water surface wetland beds (3600 and 5500 m^2, planted with *T. latifolia*; Fig 1B). The system was constructed in 2000 for the purification of the raw municipal wastewater generated by about 300 population equivalents (Mander et al., submitted, Publication VI).

2.3. Sampling and analyses

2.3.1. Gas analyses

For the measurement of N_2O, N_2, CH_4 and CO_2, two methods — the "closed chamber" ("closed soil cover box") method (Denmead and Raupach, 1993; Hutchinson and Livingston, 1993) and the He-O method (Butterbach-Bahl et al., 1997; Scholefield et al., 1997; Mander et al., 2003) were used. The latter was used especially for the measurement of N_2 fluxes.

Soil-atmosphere exchanges of greenhouse gases are usually determined directly by measuring the short-term buildup or decrease in the concentration of the gas in question in a sealed enclosure placed over the land surface. The collection system must be designed in a way that normal gaseous fluxes are not significantly affected. This method of measuring gaseous fluxes is called the "closed chamber" method.

In all experimental sites, short-term trace-gas flux measurements (approx. 1 h) were carried out using the "closed chamber" method. Gas samplers (closed chambers, cover made from PVC, height 50 cm, diameter 50 cm, volume 65 l, sealed by a water-filled ring on the soil surface, painted white to avoid heating during application) were installed in 5 replicates in various parts of the studied systems: (1) in 3 different microsites (EDGE, WET and DRY) in the Porijõgi riparian buffer zone (Fig. 1C) and in Kodijärve, (Fig. 1A) on the inlet and outlet pipes of both beds. In the hybrid wetland system in Kõo, 10 gas samplers were installed in the vertical flow filter (4 in each bed) and 15 in the horizontal flow filter (5 on two inlet pipes and 5 on an outlet pipe; Fig. 1B). At the beginning and at the end of the measuring period, gas samples were taken from the enclosures by previously evacuated gas bottles (100 ml; Augustin et al., 1998b). The trace gas concentration in the collected air was determined using the gas chromatography system (electron capture detector and flame ionization detector; Loftfield et al., 1997) in the lab of the Institute of Primary Production and Microbial Ecology, Leibniz-Centre for Agricultural Landscape and Land Use Research (ZALF) in Germany. The trace gas flux rates were calculated according to Hutchinson and Livingston (1993) from a linear change in trace

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gas concentration over time, with reference to the internal volume of the chamber and the soil area that is covered. Soil temperature and groundwater tables were measured simultaneously (Augustin et al., 1998a).

The ideal estimate of actual denitrification activity by the precise measurement of the amount of N_2O and N_2 produced is difficult to achieve in practice, because of the N_2 present in the system as a naturally high air background (Knowles, 1982). A new incubation method (the He-O method) was developed by Butterbach-Bahl et al. (1997), with which N_2 and N_2O losses by denitrification can be quantified directly.

Intact soil cores (diameter 6.8 cm, height 6 cm) for use with the He-O method were taken from the topsoil (0–10 cm) at gas sampler (closed chamber) sites after gas sampling was completed. Soil samples were weighed, kept at low temperature (4^oC), and transported to the laboratory of ZALF, Germany. At the laboratory, the intact soil cores were placed in special stainless steel incubation vessels that could be closed gas-tight to the surroundings. The soil cores within the incubation vessels were kept at the temperatures observed in the field. For the determination of actual emission rates, the following procedures were used: 1) removal of N_2 by 3 subsequent slight evacuation/flushing cycles with the artificial gas mixture (21.3% O_2, 78.6% He, 337 ppm CO_2, 374 ppb N_2O, 1882 ppb CH_4 and approx. 5 ppm N_2); 2) establishing a new flow equilibrium by continuously flushing the vessel headspace with the artificial gas mixture at 10 ml per minute for 12 hours; 3) measuring the N_2 and the greenhouse gas concentrations in the continuous gas flow (start value); 4) closing the incubation headspace for one hour to accumulate the emission of N_2 and the greenhouse gases; 5) re-measuring the gas concentrations in the incubation headspace (final value); 6) taking the difference in the gas concentrations (final accumulation value minus start continuous flow value) as the basis for the calculation of the emission rates (Butterbach-Bahl et al., 1997; Loftfield et al., 1997). Simultaneously, the NH_4-N and NO_3-N concentration in soil samples was analysed using the Kjeldahl method (APHA, 1989).

2.3.2. Water analyses

In the Porijõgi riparian alder stand, shallow groundwater samples from the upper aquifer were collected once to twice a month from piezometers installed on the borders of plant communities. Filtered water samples were analysed for $NH4^+$-N, NO_2^--N, NO_3^--N total Kjeldahl-N, PO_4^{3-}-P, total P, SO_4^{2-}, Fe, Ca^{2+} in the Laboratory of Plant Biochemistry of the Estonian Agricultural University following standard methods for the examination of water and wastewater quality (APHA, 1989).

In the Kodijärve HSSF CW, water samples were taken monthly since 1997 from the inflows and outflows of both beds, and were analysed for BOD_7, $NH4^+$-N, NO_2^--N, NO_3^--N total N, PO_4^{3-} and total P in the lab of Tartu Environmental Research Ltd (APHA, 1989).

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2.3.3. Statistical analyses

The statistical analysis was carried out using Statistica 6.0 (StatSoft Inc.). The normality of variables was checked using the Kolmogorov-Smirnov, Lilliefors', and Shapiro-Wilk's tests. In most cases, for gas analyses the distribution differed from the normal, hence non-parametric tests were performed. To check the significance of differences between the gas emission rates at different sites, the Wilcoxon Matched Pairs Test and the Mann-Whitney U-Test were used. In addition, the Duncan test was performed for the multiple comparison of the mean values of gas emission rates between the study sites. The Spearman Rank Order Correlation analysis of the relation between different parameters was performed. The level of significance of $\alpha=0.05$ was accepted in all cases.

3. RESULTS AND DISCUSSION

3.1. Trace gas emissions from alder stand and constructed wetlands

For the measurement of N_2O, CH_4 and CO_2 fluxes the "closed chamber" method was used, while N_2 fluxes were measured with the He-O method.

3.1.1. Nitrous oxide and dinitrogen flux rates

The average nitrous oxide flux from the Porijõgi riparian alder stand ranged from -3.3 to 180 μg N_2O-N m^{-2} h^{-1}. The highest values were registered in January and March 2003 from the WET microsite, during the rest of the study period N_2O-emission rates were relatively low (Fig. 2A; Teiter and Mander, 2005). Likewise, the results of some other investigations demonstrate that N_2O emission does not clearly depend on soil temperature, and the release of this gas from the soil in cold periods can be as high or even higher in winter as in summer (Augustin et al., 1996; Fey et al., 1999). Inside the Porijõgi riparian alder stand, the WET microsite emitted significantly more N_2O than the EDGE and DRY microsites (average values 30, 9 and 8 μg N_2O-N m^{-2} h^{-1} respectively (Fig. 3A; Teiter and Mander, 2005).

The average N_2 flux from the Porijõgi riparian alder stand varied from 0.02 to 17.38 mg N_2-N m^{-2} h^{-1} (Fig. 2B), the difference between N_2 and N_2O fluxes in the Porijõgi study site was 150–700 times (Teiter and Mander, 2005).

The average flux of N_2O from the Kodijärve HSSF CW ranged from 27 to 370 μg N_2O-N m^{-2} h^{-1} (Fig. 2A). According to the Wilcoxon Matched Pairs Test, significant differences were found in average N_2O fluxes between the microsites: microsites above the inflow pipes emitted approx. 10 times more N_2O-N than microsites above the outflow pipes (Fig. 3A). A significant correlation was found between the N_2O flux from all replicate chambers and the water table in both beds. An elevated water table (lower depth in cm) increased the nitrous oxide flux, especially above the inlet pipes of both beds. The average dinitrogen flux from the microsites in Kodijärve was 2–3 magnitudes higher than the N_2O flux, ranging from 0.2 to 130 mg N_2-N m^{-2} h^{-1} (Fig. 2B; Teiter and Mander 2005; Mander et al., 2003).
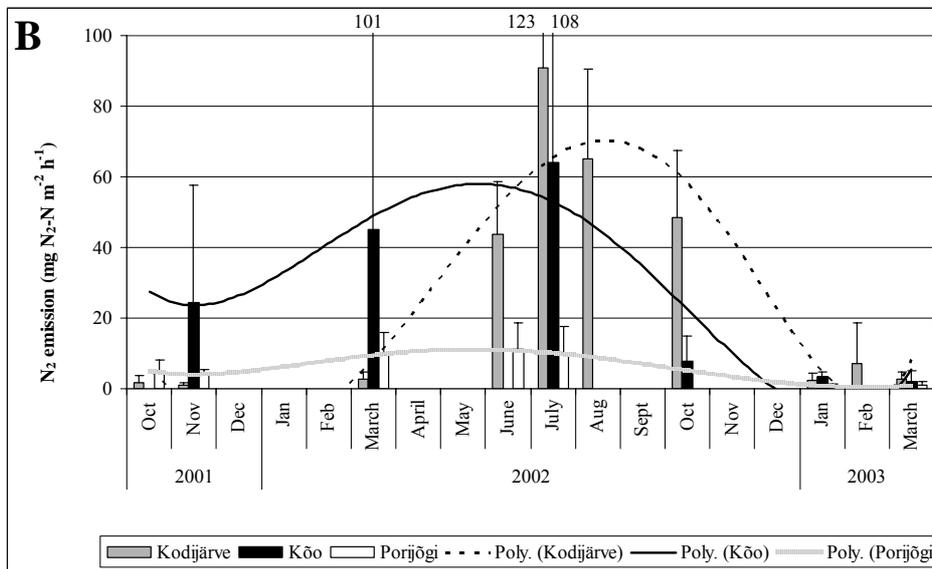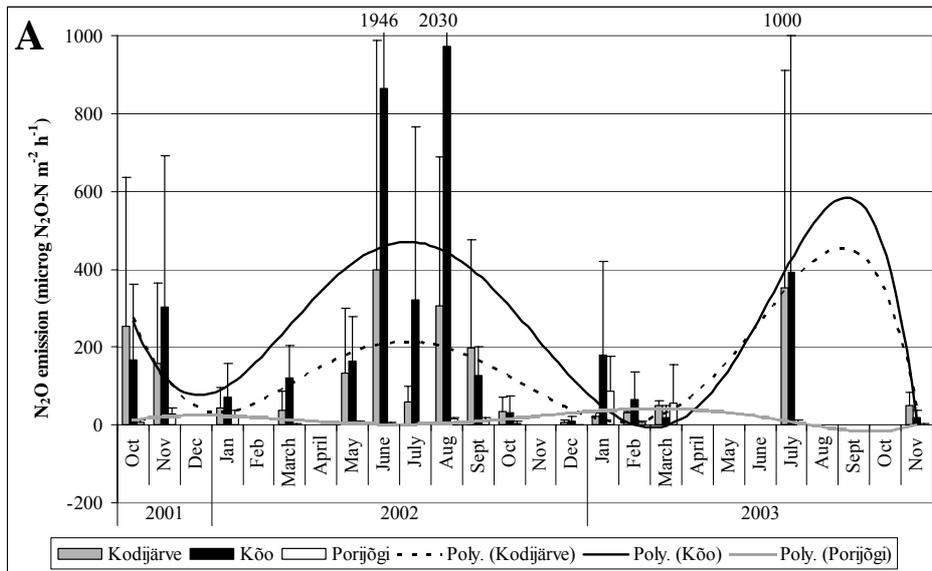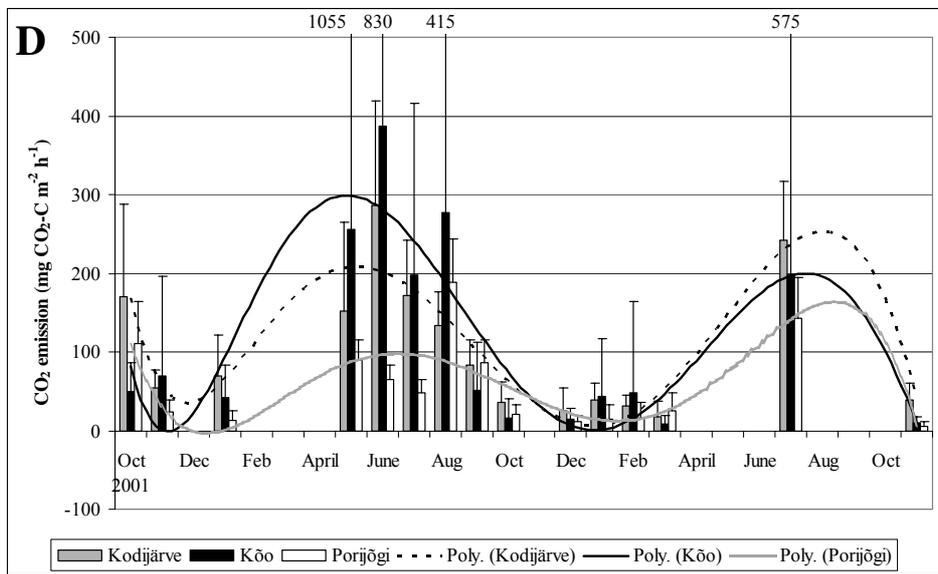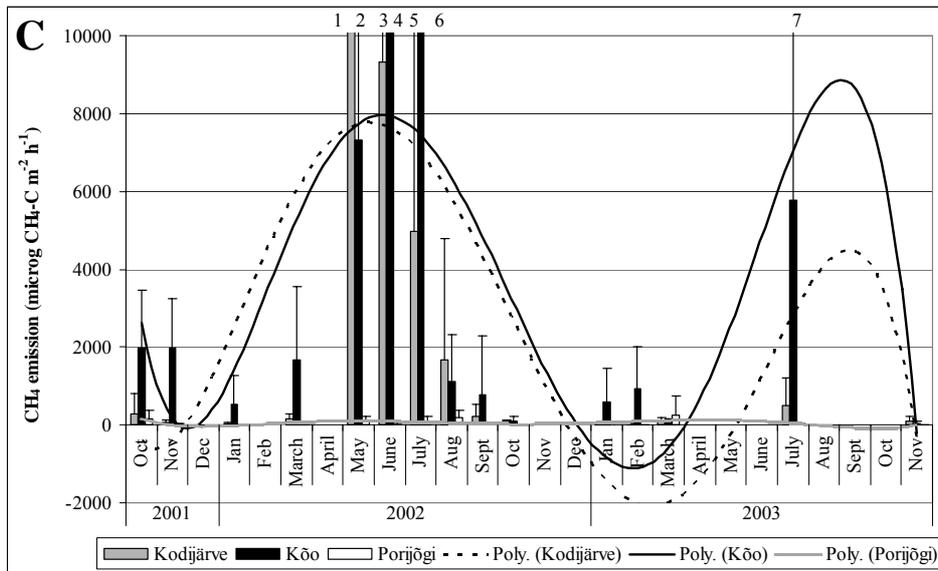
Figure 2. (continued)

Figure 2. Temporal variation of emission rates of nitrous oxide (A), dinitrogen (B), methane (C), and carbon dioxide (D; average±SD) from the Kodijärve HSSF CW, Kõo hybrid wetland system and the Porijõgi riparian grey alder stand, averaged over all sampling sites. For better visualization, polynomial curves are added. Hidden values in part C − 1: 21890±43570; 2: 18110; 3: 27425; 4: 14020±17920; 5: 1030; 6: 14410±14290; 7: 17570.

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The average flux of N_2O from the Kõo hybrid CW ranged from 72 to 500 µg N_2O-N m^{-2} h^{-1}. The VSSF beds emitted more nitrous oxide than the HSSF bed, although the differences were not significant. In Kõo, a significantly lower N_2 emission was found from the microsite above the outflow pipe of the HSSF bed (Fig. 3B). In the Kõo hybrid CW, the variation of N_2 emission was 0.3 to 99 mg N_2-N m^{-2} h^{-1} (Fig. 2B), and the difference between the N_2 and N_2O fluxes was 20–750 times (Teiter and Mander, 2005).

3.1.2. Methane flux rates

The Porijõgi grey alder forest emitted slight amounts of methane. The average CH_4 emission varied from 0.1–29 to 1.2–265 µg CH_4-C m^{-2} h^{-1} in winter and summer respectively (Fig. 2C). The WET microsite showed significantly higher (up to 396 µg CH_4-C m^{-2} h^{-1}) CH_4 emission values than the EDGE and DRY sites (Fig. 3C). This can be explained by the very high water table (0–10 cm below the soil surface) in the WET microsite (Table 1). The CH_4 flux was significantly lower than from the VSSF beds and the inflow microsites of HSSF CWs (Teiter et al., accepted, Publication I; Teiter and Mander, 2005).

The average methane emission from the microsites in the Kodijärve HSSF CW and Kõo hybrid CW ranged from 30 to 9715 and from 770 to 5540 µg CH_4-C m^{-2} h^{-1} respectively (Fig. 2C). According to the Wilcoxon Matched Pairs Test, significantly more methane was released from the microsites situated above the inlet pipes. This is consistent with the significant rank correlation between water table depth and CH_4 flux (Spearman R= −0.37). Likewise, this relationship has on many occasions been mentioned in other studies on wetlands (Boon et al., 1997, Cao et al., 1998). Also, methane release is positively influenced by suspended solids, NH_4-N, total N, PO_4-P and total P, and BOD_7 levels in wastewater. At the same time, a significant negative rank correlation was found between CH_4 flux and the NO_2-N and NO_3-N concentrations in water (Mander et al., 2003).

The CH_4 emission rates from Kodijärve and Kõo CWs are about 2 magnitudes higher than those found in natural boreal wetlands (MacDonald et al., 1998), 2–3 times higher than those reported on re-flooded fens (Augustin et al., 1996) or constructed *Typha latifolia* wetlands (Wild et al., 2002), but 2.5–3 times lower than on fertilised wet grasslands on peat soils in the Netherlands (Van den Pol-Van Dasselaar et al., 1999), up to 4 times lower than on floodplain wetlands (Boon et al., 1997), and up to 5 times lower than that observed in FWSW-s for wastewater treatment (Tai et al., 2002).
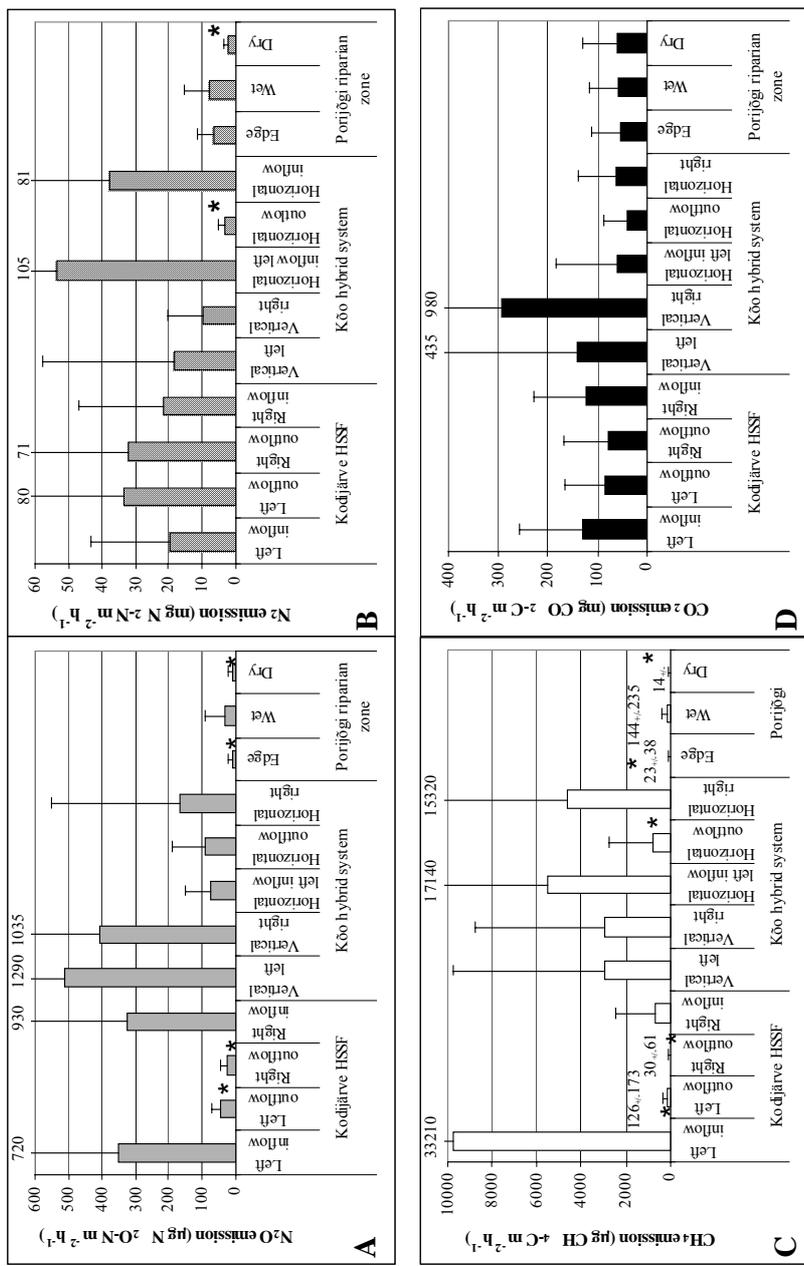
Figure 3. Emission rates of nitrous oxide (A), dinitrogen (B), methane (C) and carbon dioxide (D; average±SD) from sampling sites in the Kodijärve HSSF CW, Kõo hybrid wetland system and the Porijõgi riparian grey alder forest. * – significantly differing value ($p<0.05$) with at least two other microsites according to the Wilcoxon Matched Pairs Test. For the locations of sampling sites, see Fig. 1.

3.1.3. Carbon dioxide flux rates

In this study, CO_2 emission is not connected with fluxes related to plant photosynthesis. Therefore, only data for cold periods can be considered as losses to the atmosphere. However, some studies on C sequestration in wetlands and forest ecosystems (Butnor et al., 2003) allow one to estimate that about 50% of CO_2 released during soil respiration in the vegetation period cycles back to the atmosphere. In comparison with other gases measured, the CO_2 flux from soils showed the lowest spatial variation. Slightly higher CO_2 release was found from the microsites in the VSSF beds and above the inflow pipes of HSSF CWs (140–290 and 61–130 mg CO_2-C m^{-2} h^{-1} respectively), although these differences were not significant (Fig. 3D).

In the Porijõgi riparian grey alder stand, only the CO_2 emission varied in accordance with variations in water and air temperature. The average CO_2 emission varied from 13.6 mg CO_2-C m^{-2} h^{-1} in January to 187.8 mg CO_2-C m^{-2} h^{-1} in August (Fig. 2D).

In Kodijärve HSSF CW, the average CO_2 flux rate varied from 8–386 mg CO_2-C m^{-2} h^{-1}, and in Kõo the variation of CO_2 flux rate was 5–1120 CO_2-C m^{-2} h^{-1} (Fig. 2D).

In CWs, a clear relation was observed between the BOD_7 value of wastewater and the average CO_2 release from the filter material.

In the Kodijärve HSSF filter and in both the VSSF and HSSF parts of the Kõo hybrid CW, a significant ($p<0.05$) Spearman rank correlation was found between the fluxes of CO_2-C and emissions of N_2O, N_2 and CH_4. Likewise, the N_2O and CH_4 flux was strongly correlated in these systems. The strong relationship found between the gas fluxes from different study sites indicates the temperature dependence or occasional character of emissions. On the other hand, no significant correlation was found between the gas emissions within the Porijõgi riparian alder forest (Teiter and Mander, 2005). The last finding is probably related to the relatively high carbon storage in this riparian soil (4–5.3%; Mander et al., 1997). In filter beds of CWs, carbon can become limited due to intensive mineralization, which is reflected in the correlation between the gaseous N and C fluxes. Similar trends have been reported by Paludan and Blicher-Mathiesen (1996) for a Danish freshwater wetland, where high NO_3^- loading resulted in an accelerated loss of gaseous C.

3.2. Global warming potential of systems analysed

The global warming potential of the studied systems was calculated by converting the fluxes of N_2O and CH_4 into CO_2 equivalents. The conversion of the flux rates into CO_2 equivalents is given with 296 for N_2O and 23 for CH_4, over a time horizon of 100 years (IPCC, 2001).

The cumulative emission of N_2O and CH_4 in the riparian alder forest in Porijõgi was 0.4 ± 1.0 and 0.1 ± 0.30 t CO_2 eq ha^{-1} $year^{-1}$ respectively, whereas the CO_2-C flux was 3.5 ± 3.7 t ha^{-1} $year^{-1}$ (Fig. 4; Teiter and Mander, 2005).
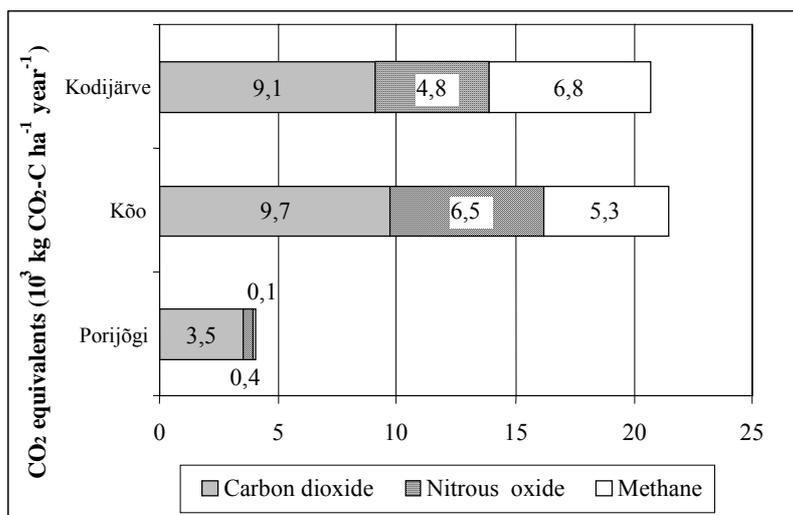

Figure 4. Proportion of major greenhouse gases from Kodijärve and Kõo constructed wetlands and Porijõgi riparian study site presented as average CO_2 equivalent values.

Both Kodijärve HSSF CW and Kõo hybrid CW emit remarkable amounts of CO_2-C, N_2O-N and CH_4-C: 9.1 ± 6.4, 4.8 ± 9.8 and 6.8 ± 16.2 t CO_2 eq ha^{-1} $year^{-1}$ in Kodijärve, and 9.7 ± 20.2, 6.5 ± 13.0 and 5.3 ± 24.7 t CO_2 eq ha^{-1} $year^{-1}$ in Kõo respectively (Fig. 4; Mander et al., submitted, Publication VI).

The emission levels of carbon dioxide and nitrous oxide in Kõo exceeded the corresponding values in Kodijärve, which is probably due to the relatively high loading of the vertical flow system (only two beds of 64 m^2 for about 300 PE). In Kodijärve the nominal loading is only 20–40 PE per 312.5 m^2. When properly functioning, however, the vertical flow system can have a relatively small area, although this seems to enhance N_2O emission. Regarding CH_4 flux, it is crucial to avoid clogging both vertical flow and horizontal flow filters: this might help in the case of a higher N_2 flux and correspondingly lower N_2O flux; however, it significantly increases methane emissions. Sometimes such clogging took place in both CWs studied, which probably led to high CH_4 emission values (Fig. 3C; Teiter and Mander, 2005).

The results of other investigations using similar measuring techniques have revealed somewhat lower emission values than in this research. Wild et al. (2002) found a GWP of FSSWs (*Typha* beds) for wastewater treatment in Bavaria, Germany from 136 to 5332 kg CO_2 eq ha^{-1} $year^{-1}$. However, the initial loading in the German system was 10–50 times lower than in the Kodijärve

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HSSF CW. On the other hand, Augustin et al. (1996), found a GWP of −31 to 7562 kg CO_2 eq ha^{-1} $year^{-1}$ for minerotrophic mires with a naturally high groundwater level and values of 3106 to 7562 kg CO_2 eq ha^{-1} $year^{-1}$ for strongly drained fens, taking into account natural CO_2 fluxes (Mander et al., 2003).

When comparing the greenhouse potential of CH_4 and N_2O over a long time scale (100–500 years), one can speculate that due to the short adjustment time for CH_4 in the atmosphere (8.4 years; IPCC, 2001), the radiative forcing of CH_4 will fall relative to CO_2 (Whiting and Chanton, 2001). N_2O, with its atmospheric lifespan of about 120 years and global warming potential value of 296, however, has a less predictable impact. Therefore, further investigations should concentrate on the factors that regulate N_2O and N_2 emission rates from CWs (Teiter and Mander, 2005).

However, when considering the limited area of existing CWs (about 10,000 around the world, of about 5000 ha in the beginning of the 1990s; (Kadlec and Knight, 1996) and even their potential area, which may be 3–5 magnitudes higher, we still get far less territory than all of the agricultural fields and natural wetlands that sometimes have higher GWP (Mander et al., 2003).

3.3. Nitrogen budgets of alder stand and constructed wetland

Water, soil and phytomass sampling and analyses for compiling nitrogen budgets are described by Mander et al., 2005a and Mander et al., submitted, Publication VI.

3.3.1. Nitrogen budget of Porijõgi grey alder stand

The total N demand in the Porijõgi grey alder stand was 285.3 kg N ha^{-1} $year^{-1}$; 81.9 kg N ha^{-1} $year^{-1}$ was transformed and assimilated by plants (Table 2). Total inputs, which consist of atmospheric deposition, subsurface and overland flow, and symbiotic N_2 fixation, are 216 kg N ha^{-1} $year^{-1}$. The non-symbiotic N_2 fixation was estimated to be negligible. Nitrogen output into groundwater and streams was 13.2 kg N ha^{-1} $year^{-1}$. The estimated N_2-N emission was 51.2 kg N ha^{-1} $year^{-1}$ (N_2O emission was negligible). In a 14-year-old grey alder stand in Porijõgi, the symbiotic N_2 fixation is higher than the gaseous N emissions (Mander et al., 2005a; Mander et al., submitted, Publication IV).

To compare nitrogen fluxes in different-aged riparian grey alder forests, Mander et al. (2005a) compiled the nitrogen budget of a 40-year-old grey alder forest in Viiratsi, southern Estonia. Inputs were significantly higher in the Porijõgi riparian alder forest due to the higher symbiotic fixation efficiency in the younger grey alder stand in the Porijõgi test area. Gaseous N-losses in Viiratsi were 48.0 kg N ha^{-1} $year^{-1}$. The Viiratsi grey alder stand was estimated to be a source of gaseous nitrogen. Many signs (higher gaseous N emissions,

less symbiotic N_2 fixation, higher net mineralization rates and less immobilization, significant increase in the proportion of understorey herbaceous plants in the budget of plant N) indicate that the buffering capacity in the older alder stand is decreasing. One can assume that 30–40 years is the maximal age for grey alder stands to provide optimal buffering capacity. Therefore these riparian ecosystems should be managed by regeneration cutting to keep their nutrient removal rate high (Mander et al., 2005a).

Table 2. Yearly nitrogen budget of Porijõgi grey alder stand (kg ha^{-1} year^{-1}; Mander et al., 2005a).

Flux or pool	
Inputs	216.6
Transformed and assimilated by plants	81.9
Litter	132.0
Trees**	103.7
Herbs	28.3
Outputs	64.9
Subsurface and overland flow	13.2
Gaseous losses*	51.7
(denitrification and nitrification) N_2O	0.5
N_2	51.2
Demand	285.3
Soil pool (t ha^{-1})	192.0

* N_2O is emitted from both denitrification and nitrification processes (Kester et al., 1997)
**   Above-ground litter

3.3.2. Nitrogen budget of Kodijärve subsurface flow constructed wetland

In the Kodijärve HSSF CW, the average annual N removal from the system was 22–25.9 kg N. The most important flux of the N budget was the N_2-N emission (11.5 kg or 52% in 2001 and 13.2 kg or 51% in 2002), followed by plant assimilation (2.3 and 1.9 kg in above-ground and below-ground phytomass in 2001 and 5.9 and 4.2 kg N year^{-1} in 2002 respectively). Microbial immobilization (2.2 and 1.0 kg N year^{-1} in 2001 and 2002 respectively), as a temporary pool, can be considered together with N accumulation in the soil (3.7 kg N year^{-1} in 2001 and 1.1 kg N year^{-1} in 2002). However, the share of these two fluxes together decreased from 27% in 2001 to 8.1% in 2002. The emission of the dangerous greenhouse gas N_2O was low: 0.4 and 0.5 kg N_2O-N year^{-1} in 2001 and 2002 correspondingly (Mander et al., submitted, Publication IV; Mander et al., submitted, Publication VI).

4. CONCLUSIONS

Current research is one of the first studies in the world where the emission rates of all three major greenhouse gases (N_2O, CO_2 and CH_4) from subsurface flow constructed wetlands were measured. Furthermore, this research is probably the first one where the N_2 fluxes from subsurface flow constructed wetlands and riparian buffer zone was measured using the He-O method.

Emissions of nitrous oxide, dinitrogen and methane from CWs showed higher values than those from the riparian grey alder forest, whereas carbon dioxide fluxes did not differ significantly. In CWs, emission rates of N_2O-N, N_2-N, CH_4-C and CO_2-C ranged from 1 to 2600 µg m^{-2} h^{-1}, 0.2–130 mg m^{-2} h^{-1}, 1.7 to 87200 µg m^{-2} h^{-1} and –6.1 to 1050 mg m^{-2} h^{-1} respectively. In the riparian grey alder forest these values were –3.3 to 180 µg m^{-2} h^{-1}, 0.02–17.38 mg m^{-2} h^{-1}, –5.9 to 805 µg m^{-2} h^{-1} and –3.9 to 290 mg m^{-2} h^{-1} respectively.

The release of all gases studied was significantly higher during the warmer period, although no significant correlation was found between N_2O flux and soil/water temperature. Like purification performance, gaseous emissions in spring and early summer were significantly lower than in autumn. The most intensive flux of N_2O and CH_4 was observed in chambers installed above the inflow pipes of horizontal flow beds. The vertical flow wetland did emit significantly more N_2O than the horizontal flow beds.

Water table increase in the horizontal flow systems may not significantly influence the efficiency of water purification, although it will increase methane emissions by a few magnitudes. Both Kodijärve HSSF CW and Kõo hybrid CW emit remarkable amounts of CO_2-C, N_2O-N and CH_4-C: 9.1 ± 6.4, 4.8 ± 9.8 and 6.8 ± 16.2 t CO_2 eq ha^{-1} year^{-1} in Kodijärve, and 9.7 ± 20.2, 6.5 ± 13.0 and 5.3 ± 24.7 t CO_2 eq ha^{-1} year^{-1} in Kõo respectively. The emission levels of carbon dioxide and nitrous oxide in Kõo exceeded the corresponding values in Kodijärve, which is probably due to the relatively high loading of the vertical flow system.

Nevertheless, even if all global domestic wastewater were treated by wetlands, their share in the trace gas emission budget would be less than 1%.

The cumulative emission of N_2O and CH_4 in the riparian alder forest in Porijõgi was significantly lower than from the CWs (0.4 ± 1.0 and 0.1 ± 0.30 t CO_2 eq ha^{-1} year^{-1} respectively), whereas the CO_2-C flux was remarkable (3.5 ± 3.7 t ha^{-1} year^{-1}).

Dinitrogen emission has been found to be the most important component in nitrogen retention from both the Porijõgi riparian grey alder forest (up to 509 kg N_2-N ha^{-1} year^{-1}) and the Kodijärve HSSF CW (up to 2500 kg N_2-N ha^{-1} year^{-1}). In the Kodijärve HSSF CW, only 1.8–1.9% of the total N removed was the dangerous greenhouse gas and ozone-layer depletion agent N_2O. Emission rates of N_2 and N_2O from normally loaded (<0.12 p.e. m^{-2}) HSSF CWs are about 400 and 10 kg N ha^{-1} year^{-1} respectively. Further implementation of the He-O method for the measurement of N_2 fluxes can give very important knowledge about nitrogen cycle in local, in regional and in global level.

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SUMMARY IN ESTONIAN

N_2O, N_2, CH_4 JA CO_2 EMISSIOON KALDAÄÄRSETES HALL-LEPIKUTES JA HEITVEEPUHASTUS-TEHISMÄRGALADES

Veekogude kaitsevööndid ja heitveepuhastus-tehismärgalad on sagedamini kasutatavad ökotehnoloogised võtted põllumajanduslikest valglatest keemilise väljakande kontrollimisel. Olulisimaks lämmastikuringet kontrollivaks protsessiks märgalaökosüsteemides ja veekoguäärsetel aladel loetakse denitrifikatsiooni. Enamikus puhvertsoonide denitrifikatsiooni käsitlevatest uurimustest on mõõdetud vaid N_2O (naerugaasi) emissiooni ja ainult väga üksikutes töödes ka N_2 vooge. Lisaks lämmastikugaaside emiteerimisele on veekoguäärsed puhverkooslused ka olulised metaani (CH_4) emissiooni kolded.

Erinevalt veekoguäärsetest puhverskooslustest on gaasiemissiooni heitveepuhastus-tehismärgaladelt märksa vähem uuritud. Enamik naerugaasi ja metaani lendumise uuringutest pärinevad vabaveelistest tehismärgaladest (madalatest veetaimestikuga tiikidest). Vaid kahes rahvusvahelise levikuga teadustöös on mõõdetud N_2O-N vooge taimestatud pinnasfiltritest.

Käesoleva uurimistöö eesmärgid on: (1) mõõta ja võrrelda N_2O, N_2, CH_4, ja CO_2 emissiooni määrasid Porijõe kaldaäärses lepikus ning Kodijärve ja Kõo olmereovett puhastavates tehismärgalades, kasutades nn suletud kambri meetodit ja He-O meetodit; (2) teha kindlaks, millised faktorid ja millisel määral N_2O, N_2, CH_4 ja CO_2 emissioonimäärasid mõjutavad; (3) võrrelda veekoguäärsest lepikust ja tehismärgaladest pärinevate N_2O, CH_4, ja CO_2 voogude alusel nimetatud ökosüsteemide kliima globaalse soojenemise potentsiaali; (4) hinnata N_2O ja N_2 emissiooni rolli Porijõe kaldaäärse lepiku ja Kodijärve tehismärgala lämmastikubilanssides.

Porijõe uurimisala paikneb Tartu maakonnas Sirvaku külas, tegemist on 20 m laiuse kaldaäärse hall-lepikuga (*Alnus incana*). Kodijärve horisontaal-vooluline pinnasfilter asub samuti Tartu maakonnas ja on rajatud 1996. a. oktoobris ca 40 inimesega Kodijärve hooldekodu heitvee puhastamiseks. Kõo hübriidne tehismärgalasüsteem Viljandi maakonnas on rajatud 2000 aastal 300 inimese olmereovee puhastamiseks.

N_2O, N_2, CH_4, ja CO_2 keskmised väärtused Porijõe kaldaäärses lepikus varieerusid vahemikus vastavalt -3.3–180 µg N_2O-N m^{-2} h^{-1}, 0.02–17.38 mg N_2-N m^{-2} h^{-1}, -5.9–805 µg CH_4-C m^{-2} h^{-1} ja -3.9–290 mg CO_2-C m^{-2} h^{-1}. Porijõe uurimisala N_2- vood ületasid N_2O-voogusid 150–700 korda. Intensiivsemat metaaniemissiooni Porijõe proovivõtukohast MÄRG võib seletada kõrge põhjaveetaseme ja vastavalt hapnikuvaesemate tingimustega mullas.

Lämmastiku eemaldamise efektiivsus kaldaäärsetest lepikutest väheneb seoses puude/koosluse ea kasvuga. Seega tuleks kaldaäärsetes lepikutes teha uuendusraiet, et eemaldada osa toitaineid ja hoida lämmastiku eemaldamise tase kõrgel.

Gaasiemissioonide keskmised väärtused tehismärgaladest varieerusid vahemikus vastavalt 1–2600 µg N_2O-N m^{-2} h^{-1}, 0.2–130 mg N_2-N m^{-2} h^{-1}, 1.7–87200 µg CH_4-C m^{-2} h^{-1} ja –6.1–1050 mg CO_2-C m^{-2} h^{-1}. Duncani testi järgi olid gaasiemissioonid soojemal perioodil oluliselt kõrgemad kui külmal ajal, ehkki N_2O voo ning mulla- ja veetemperatuuri vahel ei leitud statistiliselt olulist korrelatsiooni. Sarnaselt puhastusefektiivsusele olid ka gaasiemissioonid kevadel statistiliselt oluliselt madalamad kui sügisel. N_2O ja CH_4 vood olid horisontaalvooluliste filtrite sissevoolutorude kohal oluliselt intensiivsemad kui väljavoolutorude kohal.

Porijõe kaldaäärsest lepikust oli N_2O ja CH_4 pikaajaline emissioon vastavalt 0.4 ± 1.0 ja 0.1 ± 0.30 t CO_2 ekv ha^{-1} a^{-1}. Võrreldes kaldaäärse lepikuga olid tehismärgalade vastavad näitajad oluliselt kõrgemad: 4.8 ± 9.8 ja 6.8 ± 16.2 t CO_2 ekv ha^{-1} a^{-1} Kodijärvel ning 6.5 ± 13.0 ja 5.3 ± 24.7 t CO_2 ekv ha^{-1} a^{-1} Kõos. Vaatamata N_2O ja CH_4 kõrgetele voogudele tehismärgaladest ei ole viimaste osakaal globaalse soojenemise aspektist oluline. Isegi kui kogu kommunaalheitvesi puhastataks tehismärgalades, vastutaksid need globaalse kasvuhooneefekti tekitamisel ikkagi vähema kui 1% ulatuses.

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PUBLICATIONS

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