DISSERTATIONES TECHNOLOGIAE CIRCUMIECTORUM UNIVERSITATIS TARTUENSIS

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Removal of organic material, nitrogen and phosphorus from wastewater in hybrid subsurface flow constructed wetlands



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ORIGINAL PUBLICATIONS

- Noorvee, A., Põldvere, E. and Mander, Ü. 2007. The effect of pre-aeration on the purification processes in the long-term performance of a horizontal subsurface flow constructed wetland. Science of the Total Environment, 380, 229–236.
- II. Põldvere, E., Karabelnik, K., Noorvee, A., Maddison, M., Nurk, K., Zaytsev, I. and Mander, Ü. 2008. Hybrid constructed wetlands for wastewater treatment in Estonia. An overview of a pilot project. In: Billore, S., Dass, P. and Vymazal, J. (eds.) Proceedings of the International Conference on Wetland Systems Technology in Water Pollution Control. International Water Association. Indore (India), pp. 226–234.
- III. Põldvere, E., Karabelnik, K., Noorvee, A., Maddison, M., Nurk, K., Zaytsev, I. and Mander, Ü. 2009. Improving wastewater effluent filtration by changing flow regimes – Investigations in two cold climates pilot scale systems. Ecological Engineering, 35, 193–203.
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Author's contribution

Publication I: The author is partly responsible for the fieldwork and data collection (about 50%), and for writing the manuscript (25%).

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ABBREVIATIONS

А	_	average
ANN	_	Artificial Neural Networks
AWP	_	activated sludge wastewater treatment plants
BOD_7	_	biological oxygen demand (7 days)
CL	_	crushed limestone
COD _{Cr}	_	chemical oxygen demand (dichromic)
CW	_	constructed wetland
Fe _{tot}	_	total iron
FS	_	filter system
FWS	_	free water surface
HELCOM	_	Helsinki Commission
HLR	_	hydraulic loading rate
HSSF	_	horizontal subsurface flow
K1, K2, K3	_	long-term working periods from January 1997 to May 2008
Lr	_	mass loading rate
LWA	_	light weight aggregates
Mr	_	mass removal rate
NH ₄ –N	_	ammonium nitrogen
NO ₂ -N	_	nitrite nitrogen
NO ₃ -N	_	nitrate nitrogen
NPP	_	Net Primary Production
N _{tot}	_	total nitrogen
Р	_	phosphorus
pe	_	person equivalents
PE	_	purification efficiency
pН	_	measure of the acidity or basicity of a solution
PO ₄ -P	_	phosphate phosphorus
P _{tot}	_	total phosphorus
Q	_	discharge
S	—	standard deviation
SO_4	_	sulphate ions
TSS or SS	_	total suspended solids or suspended solids
VSSF	_	vertical subsurface flow
WB	_	water body
WC	_	water closet
yr	-	year

ABSTRACT

Constructed wetlands for the treatment of different types of wastewater are acquiring popularity, but in cold climates they are often over-designed, which prevents them from becoming as attractive as they could be.

In this PhD dissertation we assess the removal efficiency of organic material (after BOD_7 and COD_{cr}), nitrogen (after Total N (N_{tot})) and phosphorus (after Total (P_{tot})) from wastewater in the Kodjärve vertical subsurface flow (VSSF) and horizontal subsurface flow (HSSF) constructed wetland (CW) and in the Nõo and Rämsi VSSF and HSSF filter systems (FS). The Kodijärve and Nõo systems use domestic wastewater whereas the Rämsi system treats farm wastewater.

The Kodijärve CW, established in 1996, has been reconstructed twice, in 2002 when a VSSF filter was added before HSSF beds and in 2005 when filter media of HSSF beds was changed by light weight aggregates (LWA) and created the possibility to re-circulate the wastewater from the outflow well to the inflow well. Kodijärve full-scale CW performance during three long-term operation periods (K1 from January 1997 to June 2002, K2 from October 2002 to April 2005 and K3 from November 2005 to May 2008) are analysed. The VSSF in Kodijärve CW has remarkably improved aerobic conditions in both beds of the HSSF. All of the water quality indicators (dissolved oxygen, total suspended solids, biological oxygen demand, ammonia nitrogen, nitrite nitrogen, total nitrogen and total iron) improved after the construction of the VSSF filter.

The pilot scale experimental filter systems filled with LWA or crushed limestone (CL) and operated under different flow regimes in Nõo and Rämsi were established in summer 2005. The possibility of wastewater re-circulation from the interim and outflow well to the inflow well was also added. Those systems and the last reconstruction of the Kodijärve CW were supported by the Enterprise Estonia project on "Hybrid constructed wetlands for wastewater treatment" (2005–2007). Six different operational regimes were tested during those experiments.

We found that the re-circulation of wastewater is a good solution to improve the aeration and overall purification efficiency of CWs and FSs. The recirculation rate has to be from 100 to 300 percent of the inflowing wastewater to achieve satisfactory results in terms of effective BOD and COD removal and nitrification/denitrification. For example, the highest purification efficiencies in the Nõo SF, 99% and 82% for BOD₇ and Total N (N_{tot}) removal respectively, were achieved when the re-circulation rate of 300% was applied and in the Rämsi SF, 99% and 87% for BOD₇ and N_{tot} removal respectively, were achieved when the re-circulation rate of 300 to 600% was applied. In the Kodijärve CW the highest purification efficiencies, 98% for BOD₇ removal, were achieved when the re-circulation rate of 300% was applied and 58% for N_{tot} removal were achieved when the re-circulation rate of 150% was applied. To make N_{tot} removal more effective, it is wise to pump the wastewater into the septic tank, where more organic matter is available for denitrification. For sufficient Total P (P_{tot}) removal, it is crucial that the proper filter material for phosphorous removal via adsorption or precipitation be used. Another possibility is to use chemical precipitation inside the septic tank, which turned out to be the most effective way.

I. INTRODUCTION

Constructed wetlands (CW) and filter systems (FS) of various types are widely used worldwide (Kadlec and Knight, 1996). CWs and FSs are alternative wastewater treatment systems for activated sludge wastewater treatment plants (AWP) and also for packaged wastewater treatment systems.

For wastewater treatment purposes the CW were firstly carried out in the early 1950s by Dr. Käthe Seidel in Germany (Seidel, 1953). The first full scale free water surface flow wetland systems (FSW) established in the late 1960s. Subsurface flow systems were developed in the early 1970s (Kadlec and Knight, 1996). The subsurface flow constructed wetlands were primarily built with horizontal flow during the 1970s and 1980s. Vertical flow systems were developed during the late 1980s and 1990s because of the need for more effective removal of ammonia nitrogen from wastewater. Vertical and horizontal flow systems could be combined to complement each other in order to achieve higher treatment efficiency. In 1990s and early 2000s, vertical and horizontal flow systems were built in many European counties, because of more stringent requirements for example ammonia discharge. Most of constructed wetlands are primarily used to treat municipal and domestic wastewater but treatment of many types of industrial and agricultural wastewaters, landfill leachate etc. are also common. Different CWs are in operation in many European countries, e.g. Sweden, Denmark, Poland, Norway, Belgium, Estonia (Vymazal, 2006).

In addition to the satisfactory performance and multifunction of CWs and FSs in terms of biodiversity and landscape services (Vymazal, 2001), such systems have one significant disadvantage: in cold climates they are often overdesigned to compensate for uncertainty due to low temperatures, which raises construction and operating costs (Werker *et al.*, 2002). One possible operational method to achieve proper results without over-dimensioning CWs and FSs is to re-circulate the wastewater. The recirculation of wastewater as a procedure to enhance aeration and purification processes has been used in various pilot- and full-scale CWs and FSs for the treatment of different types of wastewater (Sun *et al.*, 2003; Green *et al.*, 2002; Brix *et al.*, 2002; Tchobanoglous and Schroeder, 1987).

Re-circulation provides additional oxygen transfer for aerobic microbial activities into the wastewater, and also enhances contact between the pollutants and microorganisms. Re-circulation has significantly enhanced the purification of landfill leachates (Connolly *et al.*, 2003; Zhao *et al.*, 2006). The values of water quality indicators, such as BOD and COD (Zhao *et al.*, 2004; Sun *et al.*, 2006; He *et al.*, 2006; Sun *et al.*, 1998; Del Bubba *et al.*, 2004), and concentrations of N_{tot} (Kantawanichkul *et al.*, 2001; Rustige and Platzer, 2001; Sun *et al.*, 2005; Arias *et al.*, 2005), NH₄-N (White, 1995; Sun *et al.*, 1998; Sun *et al.*, 2005; Zhao *et al.*, 2004; He, *et al.*, 2006; Sun *et al.*, 2006) and P_{tot}

(Farahbakhshazad and Morrison, 2003; Zhao *et al.*, 2004) have been reduced through the implementation of re-circulation.

The Kodijärve full-scale CW was established in 2006. The Kodijärve fullscale CW treats domestic wastewater that has crossed a ca 10 m³ septic tank. The first probes were taken from that CW in January 2007. The author of this PhD dissertation has been connected with the Kodijärve CW from September 1999 to the present. A detailed description of the Kodijärve full-scale CW and other specifications, for instance sampling etc., are presented in the current PhD chapter two, also in publication I (Noorvee *et al.*, 2007) and publication II (Põldvere *et al.*, 2008).

In Estonia there are almost thirty wetland treatment systems in Estonia (Tooming, 2005), among which there are different examples of this technology such as Kõo hybrid system (VSSF+HSSF+FSW) and Paistu hybrid system (VSSF+HSSF; Öövel, 2006). However, better overall purification efficiency in smaller area is a critical issue for most of these systems.

To develop compact hybrid constructed wetlands (CW), a project on "Hybrid constructed wetlands for wastewater treatment" was launched in December 2004. The project was supported by Enterprise Estonia project No. EU19215, and managed by the Department of Geography of the Institute of Ecology and Earth Sciences of the University of Tartu and the Institute of Technology of the University of Tartu. During the summer of 2005, three pilotscale experimental filter systems (FS) for the treatment of different types of wastewater were established, and one full-scale FS for the treatment of municipal wastewater was reconstructed in Southern Estonia (Noorvee et al., 2007; Publication I, Põldvere et al., 2008, 2009, 2010; Publications II-IV). The main objective was to test different flow regimes in order to determine optimal loading, design parameters, management schemes and operational regimes of FSs and CWs filled with crushed limestone (CL) and light weight aggregates (LWA) for the treatment of municipal wastewater (Põldvere et al., 2008, 2009, 2010; Publications II–IV). This PhD dissertation only observes continuous flow hybrid filters (see chapter 2), for an overview of monitored batch-operated filter see Põldvere et al., 2008, 2009 (Publications II-III). For the batch-operated filter, a patent (no EE 00752 U1) has been registered by the Estonian patent office.

According to Estonian legislation "*Regulation for wastewater discharge to water bodies and wastewater saturation into soil*" (RT I 2001, 69, 424), the limit values for the effluent from small wastewater treatment plants are presented in Table 1.1. In our studies (Noorvee *et al.*, 2007; Publication I, Põldvere *et al.*, 2008, 2009, 2010; Publications II–IV), we primarily followed regulations for a charge of 2000 to 9999 person equivalents (pe), and only for N_{tot} we used indicator for a charge of 10,000 to 14,999 pe. The aims of wastewater treatment regulations established by the Helsinki Commission (HELCOM, 2009) are presented in Table 1.2. Those regulations should have the

purpose of achieving further developments and investigations. For our studied systems we set the target values by charge ≥ 300 pe.

Table 1.1. Estonian legislation "*Regulation for wastewater discharge into water bodies and wastewater saturation into soil*" limit values for effluents from small wastewater treatment plants (RT I 2001, 69, 424).

Charge	Pollutants	Concent-	PE	Comment
		ration (mg l^{-1})	(≥, %)	
≥ 1999	BOD_7	no strict regula	tions, deper	nds on water body (WB) from
person	COD _{cr}		which efflu	ient is charged
equivalents	N _{tot}			
(pe)	P _{tot}			
2000 - 9999	BOD_7	15	90	up to 15–30 % stricter
ре	COD _{cr}	125	75	regulations could be applied,
	N _{tot}	_	_	depending on WB where
	P _{tot}	1.5	80	effluent is charged
10,000 -	BOD_7	15	90	up to 15–30 % stricter
14,999 pe	COD _{cr}	125	75	regulations could be applied,
	N _{tot}	15	70-80	depending on WB where
	P _{tot}	1	80	effluent is charged

Table 1.2. Wastewater treatment regulation objectives set by HELCOM

 (http://www.helcom.fi/, refer by the Estonian Ministry of the Environment).

Charge	Pollutants	Concent-	PE (≥, %)	Harness	Comment
		ration (mg I^{-1})		oneself (yr)	
≥300	BOD_5	20-40	80–95	2017-21	with water closet
person	COD _{cr}	150	_		(WC)-without
equivalent	N _{tot}	25	30-90		WC
(pe)	P _{tot}	5	70–90		
300 -	BOD ₅	25	80	2018	—
2000 ре	N _{tot}	35	30		
	P _{tot}	2	70		
2001 –	BOD ₅	15	80	2018	_
10,000 pe	N _{tot}	-	30		
-	P _{tot}	1	80		
10,001 –	BOD ₅	15	80	2015	_
100,000 pe	N _{tot}	15	70-80		
-	P _{tot}	0,5	90		2013 yr 1 mg l ⁻¹

I.I. Objectives

The main objective of this PhD dissertation is to assess the removal efficiency of organic material (after BOD_7 and COD_{cr}), nitrogen (after N_{tot}) and phosphorus (after N_{tot}) from wastewater in the Kodjärve VSSF and HSSF CW and in Nõo and Rämsi VSSF and HSSF FSs. The sub-objectives for achieving the main goal are:

- 1. To identify the removal aspects of organic material (after BOD₇ and COD_{cr}), nitrogen and phosphorus of one full-scale and two pilot-scale hybrid subsurface CW and FS in Estonia (publications I–IV).
- 2. To assess the importance of the main factors which permit the best results for the purification efficiency of organic material, nitrogen and phosphorus (publications I–IV).
- 3. To determine optimal schemes of CW and FS setup for effective organic material, nitrogen and phosphorus removal (publication I–IV).
- 4. To evaluate the cost of those systems in Estonia and to provide a comparison with active sludge treatment plants (publication III).

2. MATERIALS AND METHODS

2.1. Site descriptions

The current PhD dissertation mainly analyses one full scale (Kodijärve) CW and two pilot-scale experimental (Nõo and Rämsi) FSs in Southern Estonia, where the long-term mean ambient temperatures are -7.1° C in January and 16.5° C in July.

<u>The Kodijärve full-scale CW</u> (1997 and still operates today) is located near the hospital (40 person equivalents; pe) in southern Estonia. The domestic wastewater from a dual-chamber septic tank (circa 10 m³) outflow is channelled into an inflow well.

From the inflow well the wastewater was channelled by gravity flow into the inflow to the HSSF filters (Figure 2.1., section A). The HSSF filters consisted of two coarse iron-rich sand-filled beds, each $6.25 \times 25 \times 1$ m (312.5 m² in total), predominantly covered by *Phragmites australis* and *Scirpus sylvaticus* (see also Mander *et al.*, 2001). From approximately 2003, *Urtica dioica* and *Epilobium hirsutum were* dominant (Noorvee *et al.*, 2007; Publication I) in HSSF filters. There were eighteen water sampling wells in the HSSF.

Vertical subsurface flow (VSSF) filters (Figure 2.1., section B), filled with limestone (CL), with a total area of 37.4 m² were constructed between the septic tank and the HSSF filter in the summer of 2002 in order to enhance aerobic purification processes, especially nitrification. The wastewater is pumped into the VSSF ($2 \times 18.7 \text{ m}^2$) by a floater-controlled pump. Table 2.1 reports the cross-section of the VSSF (1.3 m in depth). The beds of the VSSF CW are loaded intermittently for a period of from one week to a month (depending on the sampling periodicity), while the other bed rests (Noorvee *et al.*, 2005; Noorvee *et al.*, 2007; Publication I). The resting of VSSF beds for several days prevents surface hydraulic and organic overloading, in order to avoid clogging (Weedon, 2003). The VSSF beds were not planted, but natural greenery, like *Urtica dioica*, grew there.

 Table 2.1. Cross-section of VSSF filters in Kodijärve CW, Nõo and Rämsi experimental FS (Põldvere *et al.*, 2008; Publication II).

Cross-section	Nõo right FS	Nõo left FS and both FSs	Kodijärve CW
		in Rämsi	
Upper layer	CL Ø 2–8 mm	LWA Ø 2–4 mm (Filtralite S)	CL Ø 4–8 mm
Middle layer	CL Ø 8–16 mm	LWA Ø 4–10 mm (Filtralite M)	CL Ø 5–20 mm
Bottom layer	CL Ø 12–32 mm	LWA Ø 10–20 mm (Filtralite L)	CL Ø 16–40 mm



Figure 2.1. Schematic layout of the Kodijärve CW, Estonia (A – diagram of the Kodijärve CW before reconstruction; B – Kodijärve hybrid CW after reconstruction in 2002 and 2005). The parallel systems (right-left units) are determined on the basis of the direction of wastewater flow.

After the VSSF, the wastewater is divided equally in the inflow well of the HSSF. The previous inflow pipe to the HSSF is used as an emergency tube for the occasion floater-controlled pump failed or a power outage took place. In the summer of 2005 the filter media of the HSSF were replaced by light-weight aggregates (LWA with particle size of 2–4 mm), because the phosphorus retention capacity of the HSSF filter was reaching its limit (Noorvee *et al.*, 2007; Publication I; Vohla *et al.*, 2005; Vohla, 2008). At the time of the construction work, the PVC liner of the HSSF was damaged and was used only one bed of the HSSF filters in most periods (Põldvere *et al.*, 2008; Publication II). At the end of 2007 the problem was fixed with a new Firestone pond liner. This liner is an inert material with low environmental impact, which elongates over 300% and has outstanding resistance to the harmful effects of ultraviolet radiation, ozone and other environmental conditions. The HSSH filter was planted repeatedly from 2005 to 2007 in spring time with *Phragmites australis* and *Typha latifolia L*.

Also in the summer of 2005, the re-circulating system of the wastewater from the outflow well to the inflow well was installed (Figure 1, section B). This allowed the enhancement of nitrification-denitrification (Põldvere *et al.*, 2008; Publication II).

For phosphorus removal, an experimental filter unit was installed in the outflow pipe (ditch) of the Kodijärve HSSF CW in summer 2002 was also tested by Vohla *et al.* (2005, 2007). The unit was filled with hydrated oil shale ash sediments and is still in operation. In autumn 2007 the filter unit was replaced with new, almost identical filter material.

<u>The Nõo experimental FS</u> is located near the existing activated sludge wastewater treatment plant (AWP) of Nõo village. The wastewater (domestic wastewater combined with dairy and meat industry wastewater) is pumped into the filter system before it reaches the screen of the AWP (Põldvere *et al.*, 2008 and 2009; Publication II and III). <u>The Rämsi experimental FS</u> is located near the existing pig farm. The wastewater entering the FS is obtained by separating the liquid fraction from the solid fraction of the swine slurry in a separation well (Põldvere *et al.*, 2008 and 2010; Publication II and IV).

In both systems a certain amount of wastewater is first pumped by a timeroperated pump into a 3-section septic tank (2 m³). After the septic tank, the wastewater is divided equally in the inflow well between both parallel (lefthand and right-hand) experimental systems, Figure 2.2. The Nõo vertical subsurface flow filter (VSSF) area is 2×4 m², and the area of the horizontal subsurface flow filter (HSSF) is 2×10 m². In Rämsi FS the area of the VSSF is 2×10 m² and the area of the HSSF is 2×15 m². Table 2.1. reports the crosssection of the VSSFs (0.7 m in depth). The HSSF filters are filled with a 1.0mdeep LWA fraction of 2–4 mm (Filtralite S), which is suitable for use in filters due to its high hydraulic conductivity, porosity and good insulation properties (Jenssen and Krogstad, 2003). The Nõo and Rämsi systems were not planted, because of the short test period, which was not sufficient for the proper growth of vegetation. Thus FSs do not meet the strict definition of a wetland, and we therefore used the term "LWA or CL filter system". The pilot filters were covered with 5-cm-thick foamed plastic slabs for insulation during winter.

Today the Nõo FS has been conserved, and the Rämsi FS has been disassembled. Results of the last experiments in Nõo experimental FS from August 2007 to March 2008 are reported by Kivisild (2009).



Figure 2.2. Plan view of Nõo and Rämsi FSs. The parallel systems (right-left units) are determined on the basis of the direction of wastewater flow.

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2.2. Experimental periods

Experimental periods for the Kodijärve CW, see Table 2.2., Nõo pilot scale FS and Rämsi pilot scale FS from November 2005 to December 2006 during the project "Hybrid constructed wetlands for wastewater treatment".

Period	Time	Time CW unit			
			area		
K1	Jan 1997 – Jun 2002	2 HSSF	312.5 m^2	no	
K2	Oct 2002 – Apr 2005	1 VSSF and 2 HSSF 1	331.2 m^2	no	
K3	Nov 2005 – May 2008	1 VSSF and 1 HSSF ^{1,2}	174.95 m ²	0–300% of	
				inflow	
3.1 ³	Nov 2005 – Dec 2006	1 VSSF and 1 HSSF ^{1,2}	174.95 m ²	0–300% of	
				inflow	

Table 2.2. Kodijärve full-scale CW performance during long-term operation periods

1 -Followed by experimental phosphorus filter unit – is not analysed here (see also from chapter 2.1). 2 -Filter media was changed. 3 -Sub-period, duration of project "Hybrid constructed wetlands for wastewater treatment", divided into six short experimental periods.

2.3. Waste water re-circulation regimes

The aim of our studies, which lasted from November 2005 to December 2006, was to explore the effect of effluent re-circulation on the purification efficiency (PE, %) of different type of filters systems.

Effluent re-circulation was achieved using timer-controlled pumps. The recirculation rates applied during the six test periods are presented in Table 2.3 and Table 2.4. The layouts of the re-circulation schemes are shown in Figure 2.1. and Figure 2.2.

The usage of timer-controlled pumps narrows the distinctions between natural treatment systems and conventional systems, but for the achieving of smaller FS areas with at least the same or better purification efficiency (PE, %), this is appropriate. Brix and Arias (2005) concluded that recycling also improved and stabilised the overall treatment performance of the VSSF CW system. Re-circulation is also used in the activated sludge processes, for instance to remove nitrogen (Tchobanoglous and Schroeder, 1987).

From 2007 to 2008 the following re-circulation rates were applied in Kodijärve CW: 250–300%, from interim well in Nõo left FS: 225–250%, and in Nõo right FS from interim well 300%. The Nõo FS HSSF filters and the outflow wells described in Table 2.4. were out of use in the above approach period.

 Table 2.3. Experiment's periods of various re-circulation regimes (% of hydraulic loading rate) of Kodijärve CW.

Recirculation regimes of Kodijärve CW 3 rd sub-period periods from Nov. 2005 to Dec. 2006	1	2	3	4	5	6
Outflow well, %	0	75	150	200	250	300

	Recirculation regimes of Nõo FS								
Periods from Nov. 2005	Right	Left	Right	Left					
to Dec. 2006	Interim	Interim	Outflow	Outflow					
	well, %	well, %	well, %	well, %					
1	0	0	25	35					
2	0	0	25	35					
3	75	50	0	0					
4	70	0	0	85					
5	150	0	150	300					
6	0	150	300	150					
Period	Rec	irculation regi	mes of Rämsi 🛛	FS					
	Right	Left	Right	Left					
	Interim	Interim	Outflow	Outflow					
	well, %	well, %	well, %	well, %					
1	25	30	40	20					
2	0	0	35	30					
3	55	40	40	30					
4	90	70	50	40					
5	300	0	300	600					
1									

Table 2.4. Re-circulation regimes (% of hydraulic loading rate) of Nõo and Rämsi FS.

2.4. Sampling

Water grab samples specified in Estonian and EU legislation were taken from the studied CWs and FSs as indicated in Table 2.5.

The water samples were analysed for pH, temperature, dissolved O_2 , redox potential, conductivity, total suspended solids (TSS), BOD₇, NH₄-N, NO₂-N, NO₃-N, N_{tot}, PO₄-P, P_{tot}, SO₄ and Fe_{tot}. (Noorvee *et al.*, 2007; Publication I). From November 2005 to December 2006, water samples were analyzed for pH, BOD₇, TSS, COD_{Cr}, N_{tot}, NH₄-N, NO₂-N, NO₃-N, P_{tot}, temperature, redox potential and dissolved O₂ (Põldvere *et al.*, 2008, 2009 and 2010; Publication II–IV). In comparison, the purification efficiencies and removal rates of organic matter (after BOD₇ and COD_{cr}), NH₄-N, N_{tot}, and P_{tot} were used as performance indicators.

Sampling session	System	Sampling points from	Average frequency
Jan 1997 to Jun 2002	Kodijärve	inlet and outlet of both HSSF ¹	once a month
Oct 2002 to Apr 2005	Kodijärve	inlet and outlet of VSSF and from the inlet and both outlets of HSSF wetlands and the outlet of the phosphorus removal bed ²	once a month
Nov 2005 to Dec 2006	Kodijärve, Nõo and Rämsi	the outlet of the septic tank and the outlet of VSSF and HSSF beds	once a week ³
Jan. 2007 to May 2008	Kodijärve	inlet and outlet of VSSF and from the inlet and mean outlet of HSSF wetlands	once a month
Sept. 2007 to March 2008	Nõo	before and after septic tank and outlet of VSSF	once a month

Table 2.5. Sampling sessions in Kodijärve CW, Nõo FS and Rämsi FS.

1 - water sample series were taken also from eighteen 50mm polyethylene wells that were inserted in both beds of the HSSF systems (see Figure 2.1., section A); 2 - results are not presented in this work (see Vohla *et al.*, 2005, 2007 and Vohla, 2008); 3 - the Rämsi pilot-scale CW was partly frozen during the 2^{nd} period in wintertime, and partly clogged during the 3^{rd} and 4^{th} periods.

2.5. Filter materials used in constructed wetlands and pilot filter systems

From January 1997 to April 2005, Kodijärve CW HSSF filters consisted of two coarse iron-rich sand-filled beds (Mander *et al.*, 2001). In the summer of 2002 VSSF beds filled with crushed limestone (CL) were added to Kodijärve CW. The experimental phosphorus removal filter unit, installed in the outflow pipe (ditch) of the Kodijärve HSSF CW in summer 2002, was filled with hydrated oil shale ash sediments (Vohla *et al.*, 2005, 2007 and Vohla, 2008), and those sediments were exchanged in 2007.

After November 2005, the LWA used in Kodijärve CW HSSF, Nõo and Rämsi pilot filter systems was produced from local clay mineral in Estonia (trademark name Filtralite S, M, L), and does not have the characteristics of Filtralite-P. The Estonian LWA does not have a high phosphorus sorption capacity (Jenssen and Krogstad, 2003), and is not crushed.

In one VSSF in Nõo, we used CL that had relatively poor properties (larger grain size, dusty and different solubility properties) compared to the LWA used (smaller grain size and therefore higher surface area).

2.6. Purification efficiency and mass removal rate

The purification efficiency (PE; %) of water quality indicators was calculated using the following equation (Kadlec and Knight, 1996):

$$Pe = (C_{in} - C_{out}) / C_{in} * 100$$
(1)

where:

 C_{in} = average value of inflow concentration (mg l⁻¹); C_{out} = average value of outflow concentration (mg l⁻¹).

Mass removal (Mr; $g m^{-2} d^{-1}$) was calculated using the following equation (Kadlec and Knight, 1996):

$$Mr = [(C_{in} * Q_{in}) - (C_{out} * Q_{out})]/A$$
(2)

where:

6

A = area of CW (m^2) ;

 Q_{in} and Q_{out} = average values of water discharge in inflow and outflow $(m^3 d^{-1})$;

 C_{in} and C_{out} = average values of inflow and outflow concentrations (mg l^{-1}).

2.7. Statistical analysis

Initial data were verified for normality (Kolmogorov-Smirnov, Lilliefors' and Shapiro-Wilk's tests). Since the raw data were not normally distributed, nonparametric Kruskal-Wallis ANOVA tests were carried out for the comparison of the purification efficiencies of different periods, and a non-parametric Spearman Rank Order Correlation coefficient was detected between influencing factors and purification efficiencies of BOD₇, COD_{Cr}, N_{tot}, NH₄-N and P_{tot}. Most of the mass removal and purification efficiency values were normally distributed.

Summarized recirculation rates from the outflow and interim wells were used in the correlation analysis in the case of the Kodijärve CW and Nõo and Rämsi FSs.

The level of significance of $\alpha = 0.05$ was accepted in all cases. *Statistica* 7.0 was used for the data analysis.

3. RESULTS AND DISCUSSION

In the current PhD dissertation, experimental cycles of the Kodijärve CW, Nõo FS and Rämsi FS are analysed.

Average values of the mass loading rates of the experimental period from January 1997 to May 2008 are given in Table 3.1., and from November 2005 to December 2006 in Table 3.2.

Table 3.1. Average values of the mass loading rates $(g m^{-2} d^{-1})$ in the inflow of the Kodijärve CW during working periods K1, K2, K3 from January 1997 to May 2008 (A – average; S – standard deviation).

	Average	Average mass loading rates in the inflow of Kodijärve CW (g m ⁻² d ⁻¹)												
Period	BOD ₇		NH ₄ _N		N _{tot}		P _{tot}							
	Α	S	Α	S	Α	S	Α	S						
K1	1.3	1.3	0.7	0.6	0.8	0.7	0.2	0.2						
K2	2.5	1.6	1.8	1.0	2.0	1.1	0.3	0.1						
K3	2.8	1.3	2.0	0.8	2.3	1.0	0.4	0.3						

Table 3.2. Average inflow values of mass loading rates (g $m^{-2} d^{-1}$; A – average; S – standard deviation) during experimental periods from November 2005 to December 2006. For explanation of periods see chapter 2.2.

		Mass loading rates (g m ⁻² d ⁻¹)											
Period	System	BOD ₇		CO	D _{Cr}	N _{tot}	t	P _{tot}					
		Α	S	Α	S	Α	S	Α	S				
1	Nõo	19.2	5.8	35.7	7.3	3.8	1.3	1.1	0.2				
05 Nov-	Rämsi	88.3	31.9	112.6	33.2	8.1	2.1	3.1	1.3				
05 Dec	Kodijärve	2.4	0.7	4.7	0.9	2.1	0.2	0.4	0.0				
2	Nõo	11.2	1.8	19.8	3.0	1.9	0.5	0.5	0.1				
06 Jan-	Rämsi	22.8	1.1	32.3	1.0	2.1	0.6	0.6	0.2				
06 Mar	Kodijärve	4.0	2.2	8.3	3.3	2.5	0.8	0.5	0.1				
3	Nõo	9.2	4.5	16.7	10.8	1.1	0.4	0.4	0.2				
06 Mar-	Rämsi	15.4	8.4	22.0	11.1	1.2	0.6	0.5	0.2				
06 May	Kodijärve	3.7	1.2	7.1	1.5	2.4	0.7	0.7	0.3				
4	Nõo	10.1	2.2	16.9	5.8	1.4	0.5	0.5	0.1				
06 May-	Rämsi	29.9	6.4	44.7	3.2	2.4	0.3	1.2	0.3				
06 Jul	Kodijärve	3.4	1.1	8.8	6.0	2.3	0.3	0.4	0.2				
5	Nõo	13.8	3.2	24.3	11.4	2.4	0.7	1.0	0.3				
06 Aug-	Rämsi	23.3	5.5	36.3	7.4	2.1	0.4	1.0	0.2				
06 Oct	Kodijärve	2.0	0.8	5.2	2.3	1.9	0.9	0.4	0.3				
6	Nõo	5.4	1.1	8.4	0.8	1.1	0.1	0.4	0.1				
06 Oct-	Rämsi	18.6	4.3	29.0	2.4	2.1	0.2	0.8	0.2				
06 Dec	Kodijärve	3.0	0.8	5.9	1.2	2.7	0.6	0.4	0.1				

Average values of the hydraulic loading rates, discharge (Q, $m^3 d^{-1}$), pH and water quality concentration parameters in the inflow of the studied filter systems during regimes of experiments from January 1997 to May 2008 are given in Table 3.3., and from November 2005 to December 2006 in Table 3.4.

Table 3.3. Average values of the hydraulic loading rates (HLR, mm d⁻¹; without recirculation), discharge (Q, m³ d⁻¹), pH and water quality concentration (mg Γ^{-1}) parameters in the inflow of the Kodijärve CW during working periods K1, K2, K3 from January 1997 to May 2008 (A – average; S – standard deviation).

	HLR	Q			Average water quality parameters in the inflow of Kodijärve CW (mg I ⁻¹)							
	(mm	(m^3)	р	Н	BO	\mathbf{D}_7	N	tot	NH	I ₄ –N	P _t	ot
Period	d ⁻¹)	d ⁻¹)	Α	S	Α	S	Α	S	Α	S	Α	S
K1	99.2	3.2	7.5	0.2	128.1	61.4	99.1	31.9	85.4	27.1	14.7	4.7
K2	45.5	7.3	7.5	0.1	117.4	47.7	90.4	23.0	80.4	25.8	11.6	2.1
K3	30.16	5.8	7.5	0.2	95.5	41.5	75.2	18.8	66.0	18.3	14.2	5.5

Table 3.4. Average	e values of t	he hydrau	ulic loadin	g rates (HI	R, mm d ⁻¹ ; with	out re-circulation)	, discharge (Q, m	³ d ⁻¹), pH and
water quality conc-	entration (m ₃	g l ^{-l}) par	rameters in	n the inflov	of the studied f	ilter systems duri	ng different flow	regimes (A –
average; S - standa	urd deviation) of expe	srimental po	eriod from	November 2005	o December 2006	. For an explanati	on of periods,
see chapter 2.2.								
								,

see chapte	r 2.2.														
		a m	C	d	Η	Аvеı	age wate	er quality	paramet	ters in th	e inflow	7 of FSs	and C	W (mg I	-1)
Period	System	HLK	رس ^{ع را-1}	•	ŭ	BC	$\mathbf{D}_{\mathbf{J}}$	CO	D _{Cr}	Ž	ot	NH4	Z	\mathbf{P}_{to}	
		(n mm)	(n III)	¥	2	A	S	A	S	A	S	V	S	A	S
1	Nõo	52	0.73	7.4	0.2	405	102	745	141	72	26	52	12	20	б
05 Nov-	Rämsi	16	0.4	9.9	0.8	5519	1993	7039	2073	507	132	412	123	191	80
05 Dec	Kodijärve	22	3.8	7.5	0.1	109	32	217	44	96	7	90	4	17	1
2	Nõo	26	0.37	7.6	0.2	425	69	750	112	71	20	52	15	21	4
06 Jan-	Rämsi	4	0.1	6.3	0.1	5700	265	8068	247	527	143	431	76	158	38
06 Mar	Kodijärve	30	5.3	7.5	0.2	133	72	274	111	83	27	70	23	16	ς
3	Nõo	21	0.29	7.4	0.4	446	216	808	519	55	18	47	14	21	8
06 Mar-	Rämsi	4	0.1	6.2	0.3	3838	2089	5512	2783	305	142	265	120	120	50
06 May	Kodijärve	34	9	7.6	0.4	108	34	206	4	71	20	61	12	20	8
4	Nõo	16	0.22	7	0.1	644	138	1077	369	92	33	78	39	31	6
06 May-	Rämsi	4	0.1	5.8	0.1	7483	1596	11180	789	594	70	474	42	297	66
06 Jul	Kodijärve	34	5.6	7.4	0.1	105	36	276	186	71	6	59	16	12	9
5	Nõo	21	0.3	7.2	0.3	646	150	1136	533	112	33	81	33	45	15
06 Aug-	Rämsi	5	0.13	5.7	0.2	4656	1105	7267	1471	426	89	368	76	194	34
06 Oct	Kodijärve	34	5.6	7.5	0.1	70	12	180	44	67	18	59	17	14	8
9	Nõo	14	0.2	7.2	0.2	376	75	590	59	78	10	62	11	26	5
06 Oct	Rämsi	4	0.1	6.4	0.2	4655	1078	7246	601	517	58	396	60	208	43
06 Dec	Kodijärve	40	7	7.5	0.1	74	19	148	30	67	14	56	12	11	0

3.1. Kodijärve full-scale constructed wetland

3.1.1. Performance results of Kodijärve full-scale constructed wetland system from January 1997 to May 2008

The data presented in this chapter includes three operation periods, see Table 2.2. The average wastewater inflow parameters of those three periods are presented in Table 3.1. and Table 3.3. The hydraulic loading rate was lower in the 3rd period (Table 3.3.), when average wastewater inflow was 5.8 m³ d⁻¹. In the third period one HSSF filter bed was out of use (Table 2.2.) due to technical problems, (Põldvere *et al.*, 2008; Publication II) and therefore the highest values of mass loading rates were achieved (Table 3.1. and Figure 3.1.).

Figure 3.2. and Figure 3.3. provide a review of average purification efficiency (PE) and mass removal rate (Mr) in working periods K1, K2, K3. These reveal that the implementation of the VSSF filter (in year 2002) had a positive influence on overall purification efficiency (Noorvee *et al.*, 2007; Publication I). The mass removal rate of organic matter (after BOD₇) was higher in the Kodijärve CW VSSF filter in the 2^{nd} period (K2). In the 3^{rd} period (K3) a rise in the mean P mass removal rate was observable, but this was not significant (Figure 3.3.).



Figure 3.1. Average mass loading rates (g m⁻² d⁻¹) of organic matter (after BOD₇), NH₄-N, N_{tot} and P_{tot} in the inflow of Kodijärve CW from January 1997 to May 2008 in working periods K1, K2, K3. No significant differences (p < 0.05; Kruskal-Wallis test) have been found.

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Figure 3.2. Average purification efficiencies (PE %) and mass removal rates (Mr g m⁻² d⁻¹) of organic matter (after BOD₇), NH₄-N, N_{tot} and mean outflow in Kodijärve CW VSSF filters from January 1997 to May 2008 in working periods K1, K2, K3. No significant differences (p < 0.05; Kruskal-Wallis test) have been found.



Figure 3.3. Average purification efficiencies (PE %) and mass removal rates (Mr g m⁻² d⁻¹) of organic matter (after BOD₇), NH₄-N, N_{tot} and in mean outflow of Kodijärve CW HSSF filters from January 1997 to May 2008 in working periods K1, K2, K3. No significant differences (p < 0.05; Kruskal-Wallis test) have been found.

 NH_4 N and N_{tot} average purification efficiency did not show an improvement in the 3rd period, which may be caused by the lack of organic matter to complete de-nitrification and also by space availability (Table 2.2.). According to Laber *et al.* (2003) for effective de-nitrification the ratio of COD_{cr}/N_{tot} should theoretically be at least 2.8 (ratio of C/N_{tot} 0.7), and in actual conditions it is convenient to use a ratio of COD_{cr}/N_{tot} of 6.0 (ratio of C/N_{tot} 1.5). We only carried out COD_{cr} analyses from wastewater in the 3rd working period, and those analyses we were constrained by the time limit from November 2005 to December 2006. For that period we can also refer to the ratio of COD_{cr}/N_{tot} , see Table 3.5.

Table 3.5. Average values of ratio COD_{cr}/N_{tot} for experiment's periods of various recirculation regimes (Laber *et al.*, 2003 – theoretical minimal value: 2.8; optimal value in field conditions: 6.0). For an explanation of periods, see Table 2.2.

3 rd sub-		Ratio of CO	D _{cr} /N _{tot} in the Kod	ijärve CW
period	Inflow to	Outflow from	Outflow from	Inflow with re-circ.
periods	VSSF	VSSF	HSSF	from HSSF
1	2.3	1.2	1.0	1.8
2	3.3	1.8	1.2	2.4
3	2.9	1.9	1.3	2.4
4	3.9	2.0	1.1	2.8
5	2.7	1.7	0.9	2.0
6	2.2	0.9	0.6	1.6

As appears from Table 3.6, one must employ future action to meet standards, especially for N_{tot} and P_{tot} , which are set by the regulations. Some possible ways for achieving better purification efficiency are presented in chapter 3.3.

Table 3.6. Outflow water parameters and purification efficiency of Kodijärve CW in different working periods and in accordance with the regulations in Table 1.1. and Table 1.2.

Period	Variable	Out conce ns r	tflow ntratio ng l ⁻¹	Purifi effic	ication iency %	Complies with regulations in Table 1.1	Complies with regulations in Table 1.2
		Α	S	Α	S		
K1	BOD ₇	17.5	16.2	85.7	13.8	no	yes
K2		3.1	2.3	97.3	2.4	yes	
K3		7.1	12.3	93.5	7.1	yes	
K1	N _{tot}	48.2	15.9	47.6	20.7	no	mg l^{-1} no, but %
K2		41.2	14.6	53.2	13.4	no	yes
K3		45.6	12.7	39.1	15.5	no	
K1	P _{tot}	3.2	1.7	76.4	18.9	no	yes
K2		4.2	1.3	59.1	10.6	no	mg l ⁻¹ yes,
							but % no
K3		5.9	2.1	54.5	16.8	no	no

3.1.2. Performance results of Kodijärve constructed wetland system from November 2005 to December 2006

The long long-time inflow parameters, outflow parameters of wastewater and performance summary of the Kodijärve CW system before November 2005 is presented in chapter 3.1.1 of this PhD dissertation. After establishment of the VSSF filter, we observed that the VSSF CW had remarkably improved aerobic conditions in both beds of the HSSF filter. The dissolved oxygen content increased significantly in both filter beds (p<0.001). Except for total P concentrations in the right bed and NO₃-N concentrations in the outflow of both beds, all of the water quality indicators improved after construction of the VSSF bed. No significant seasonal effects on the purification efficiency of the water quality indicators were found (Noorvee *et al.*, 2007; publication I).

In the summer of 2005 the HSSF filter media were replaced. We used lightweight aggregates (LWA), which we hoped would improve phosphorus removal results. In addition, the re-circulation of water to enhance denitrification was implemented. Since it was not possible to change the hydraulic loading in the <u>Kodijärve CW</u>, the differences in system performance could only be controlled through re-circulation (Table 2.3.). Increase of recirculation improved purification efficiency of all water quality indicators, except for N_{tot} and P_{tot} (Table 3.7.). We observed a significant improvement in BOD₇ purification efficiency, compared to the last operational period, with higher re-circulation than the first operational periods. We found a significant positive correlation between re-circulation rates and the purification efficiencies of BOD₇, COD_{Cr} and NH₄-N. A significantly positive correlation between water temperature and BOD₇ purification efficiency was also observed (Põldvere *et al.*, 2008; Publication II). Values for the ratio of COD_{cr}/N_{tot}, which could also affect the purification efficiency of N_{tot}, are presented in Table 3.5.

Mass removal rates are adduced in Table 3.8. In the Kodijärve CW system the mass loading rate depended on whether both beds of the HSSF filter were in use or only one bed. The mass removal rate approached the values of the mass loading rate (Table 3.2.) in periods with higher re-circulation.

Basically we succeeded in meeting Estonian standards only for BOD₇ and COD_{cr} values. Considering the aims of the wastewater treatment regulations established by the Helsinki Commission, we met also the limits set for BOD₇ and COD_{cr} . For N_{tot} and P_{tot} we obtained results that did not assure the demanded concentration (mg l⁻¹) in most periods, but they met the targets set for purification efficiency (%).

After December 2006, when the project "Hybrid constructed wetlands for wastewater treatment" (supported by Enterprise Estonia project No. EU19215) was finished, the average inflow parameters and system performance efficiency remained almost the same, and did not affect the overall results, which are presented in chapter 3.1.1.

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		-	,						ò				-		-	
		PE of E	$3OD_7$			PE of C	OD_{cr}			PE of	N_{tot}			PE of	$\mathbf{P}_{\mathrm{tot}}$	
	VSS	Ε	SSH	Ľ,	VSS	Ч	HSS	н	VSS	Ч	SSH	н	VSS	ГŢ	ISSH	[1]
Period	Α	S	A	S	A	S	A	S	A	\mathbf{N}	A	S	A	S	A	S
1	80.0	14.9	91.0	4.1	63.6	19.7	74.0	6.4	33.7	20.0	39.0	11.1	43.1	22.8	58.0	8.7
2	72.3	14.2	85.0	8.0	60.3	16.3	74.0	13.1	25.5	15.9	28.0	11.5	37.3	22.0	47.0	15.0
3	72.2	19.9	90.0	10.5	57.1	15.3	78.0	9.0	32.7	17.7	52.0	23.4	43.0	27.8	73.0	20.3
4	68.0	21.6	97.0	1.7	60.9	28.5	82.0	11.4	24.9	11.5	36.0	13.4	23.1	18.2	64.0	15.1
5	72.1	15.9	97.0	2.1	44.4	22.4	81.0	17.0	11.5	10.4	42.0	15.2	35.6	19.6	66.0	19.8
9	90.9	5.3	98.0	0.6	71.8	11.1	83.0	6.0	34.1	11.8	37.0	9.7	38.6	11.8	45.0	10.9

Table 3.7. Purification efficiency (PE) of organic matter (after BOD₇ and COD_{cr}), N_{iot} and P_{iot} in Kodijärve CW from November 2005 to December 2006 in periods 1–6 (mean outflow of VSSF and HSSF, A – average; S – standard deviation). For explanation see chapter 2.2.

Table 3.8. Mass removal rate (Mr) of organic matter (after BOD₇ and COD_a), N₁₀₁ and P₁₀₁ in Kodijärve CW from cycle Nov 2005 to Dec 2006 in sub-periods 1–6 (mean outflow of VSSF and HSSF, A – average; S – standard deviation). For explanation see chapter 2.2.

	•	,)				•		•		
		Mr of B	OD_7		_	Mr of C	OD_{cr}			Mr of]	Ntot			Mr of]	P_{tot}	
	VSS	Я	SSH	Ľ,	VSS	Ъ	ISSH	ĹĿ	VSS	Ч	ISSH	٢.	ISSV	ĹĿ	ISSH	ſ.,
Period	A	S	A	S	A	S	A	S	A	S	A	S	A	S	A	S
1	17.6	7.1	2.1	0.7	28.0	12.8	3.5	1.0	6.6	4.5	0.8	0.3	1.5	0.9	0.2	0.0
2	27.1	10.7	3.4	1.5	46.5	17.1	6.1	2.6	6.0	10.1	0.7	0.7	1.7	1.0	0.2	0.1
3	25.1	11.1	3.3	1.2	38.0	15.4	5.6	1.6	7.5	6.6	1.3	0.9	2.8	3.0	0.5	0.3
4	20.3	13.4	2.8	1.5	48.4	53.0	6.5	6.3	6.5	4.8	0.7	0.4	1.0	1.0	0.2	0.2
5	15.8	3.7	2.2	0.4	27.4	13.0	4.7	1.8	4.3	7.0	0.9	0.6	1.7	1.7	0.3	0.3
9	25.2	7.0	2.9	0.8	39.7	11.5	4.9	1.1	8.6	4.4	1.0	0.4	1.6	0.7	0.2	0.1

3.2. Nõo and Rämsi experimental pilot filter systems

With the decrease in wastewater and pollutant load (Table 3.2. and Table 3.4.) and the greater re-circulation of wastewater (Table 2.4.), purification efficiency increased in <u>Nõo</u> (Table 3.9.) and <u>Rämsi FSs</u> (Figure 3.10.) in most water quality indicators. For instance, the purification efficiency of BOD₇, COD_{Cr}, N_{tot} and NH₄-N increased in both the right-hand and left-hand systems of the FSs. The mass removal rate (g m⁻² d⁻¹) followed almost the same pattern as purification efficiency, see Table 3.11. and Table 3.12.

As expected, we found a significant positive correlation between the recirculation rate and purification efficiency of BOD₇, COD_{Cr} , N_{tot} and NH_4 -N in both parallel systems of <u>Nõo</u> and <u>Rämsi FS</u> (Põldvere *et al.*, 2008, 2009 and 2010; publication II, III, IV).

In <u>Nõo FS</u> the left-hand system showed a more stable purification efficiency than the right-hand filter system. Better overall purification efficiencies in the water quality parameters were noted in the 2^{nd} , 3^{rd} , 4^{th} and 5^{th} periods, and only P_{tot} purification efficiency was better in the right CL and LWA filter in the 2^{nd} , 3^{rd} and 4^{th} periods, when the re-circulation from the outflow well was smaller (Table 2.4.). The poor purification efficiency in the right parallel indicates the influence of higher re-circulation in the left bed during the investigated periods and the fact that LWA is a better filter material than crushed CaCO₃-rich limestone in VSSFs, because of its higher porosity and longer residence time in the filter.

In the left-hand system we also observed a significantly positive correlation with inflow temperature (°C) and BOD₇ purification efficiency. Table 3.13. presents values for the ratio of COD_{cr}/N_{tot} , which could also affect the purification efficiency of N_{tot} . As can be seen, the above-mentioned ratio in the HSSF filters is below the recommended ratio of 6.0 (Laber *et al.*, 2003), but in the inflow well this ratio was guaranteed. The comparison between the two parallel systems is not direct, because of the difference in the VSSF filter media (Table 2.1.).

In the left-hand filter system of the LWA filter we also found a significant negative correlation between inflow (Q) and P_{tot} purification efficiency. The significantly lower purification efficiency of P_{tot} during the 3rd operational regime in the Nõo filter system could have an effect on the reaction balances in the vertical filter, because enhanced inflow of O₂-rich water from the interim well may suppress the solubility of CO₂. Thus the average CO₂ concentration could become the limiting factor in the geochemical pathway that leads to the final sedimentation of P (Zaytsev *et al.*, 2007).

matter (after BOD ₇ and COD _{cr}), N _{tot} and P _{tot} in Nõo FS from November 2005 to December	HSSF filters; left (L) and right (R) sides are determined on the basis of wastewater flow)	on see chapter 2.2.
BOD ₇ and COD _{cr}), N _{to}	left (L) and right (R) s	r 2.2.
of organic matter (after	VSSF and HSSF filters;	r explanation see chapte
ation efficiency (PE)	-6 (mean outflow of ¹	standard deviation. For
Table 3.9. Purific	2006 in periods 1	A – average; S –

Table 3.9 2006 in pt A – averaț	Purific: eriods 1- ge; S - s	ation eff -6 (mear tandard	iciency (1 outflov deviatio	(PE) of o v of VSS n. For ex	rganic n 8F and H planatio	natter (a ISSF filt n see ch	fter BOI ers; left apter 2.2	D ₇ and C (L) and	OD _{er}), N right (R	J _{tot} and P) sides a	_{tot} in Nõ tre deter	o FS from	n Novei 1 the bas	mber 200 sis of wa	05 to De istewatei	cember flow).
				PE of	BOD_7							PE of ($OD_{\rm cr}$			
	L – J	VSSF	L - H	HSSF	R - F	HSSF	R –	VSSF	L - V	/SSF	L - F	ISSF	R - F	ISSF	R - V	SSF
Period	A	\mathbf{N}	A	S	A	S	A	S	A	S	A	S	A	S	A	\mathbf{v}
1	66.2	15.9	67.0	10.8	51.0	15.4	55.4	4.4	62.2	8.6	64.0	18.8	51.0	19.6	49.1	8.5
7	78.1	10.5	82.0	9.9	76.0	9.7	63.8	7.3	71.3	9.1	78.0	10.3	72.0	9.7	55.1	8.5
Э	89.5	7.2	89.0	8.1	87.0	11.5	77.5	11.9	82.6	8.8	86.0	6.8	84.0	11.0	71.4	16.4
4	67.3	16.1	89.0	11.6	84.0	8.7	56.0	21.3	59.9	15.7	83.0	18.3	82.0	7.3	55.7	15.8
5	90.4	8.4	98.0	1.1	95.0	3.8	87.4	8.8	83.5	11.1	94.0	5.1	92.0	9.0	79.5	17.5
9	95.3	4.4	0.66	0.3	0.66	0.5	95.7	4.7	86.7	8.3	93.0	2.8	93.0	3.0	88.4	4.2
				PE 0	${f f} {f N}_{ m tot}$							PE of	P _{tot}			
	L - J	VSSF	L - I	HSSF	R - F	HSSF	R – 1	VSSF	L - V	/SSF	L - F	ISSF	R - F	ISSF	R - V	SSF
Period	A	S	A	S	A	S	A	S	A	S	A	S	A	S	A	S
1	25.3	8.1	5.0	15.1	11.0	15.9	16.6	11.4	38.0	17.3	47.0	22.6	76.0	14.9	45.7	26.5
2	27.9	12.1	28.0	28.6	25.0	28.5	9.0	13.0	17.9	12.1	31.0	26.3	66.0	14.9	20.9	21.4
3	49.9	14.4	46.0	15.2	38.0	14.0	27.2	15.2	22.7	17.2	40.0	12.6	45.0	20.4	22.0	19.6
4	55.4	19.2	66.0	14.1	51.0	18.2	50.4	18.1	16.8	19.0	67.0	31.9	75.0	29.9	22.4	19.3
5	67.7	18.3	80.0	8.2	74.0	11.1	69.6	7.0	52.7	21.2	76.0	15.9	73.0	13.4	53.2	20.3
9	69.4	3.3	80.0	2.7	82.0	2.7	66.0	3.5	44.5	16.2	69.0	13.7	61.0	9.2	35.5	8.1

Decembei	: 2006 in	periods	I6. A -	- average	e; S – sta	andard de	eviation.	. For exp	lanation	see Tab	le 3.9.					
				PE of	BOD_7							PE of C	OD_{cr}			
	L – J	/SSF	L - F	ISSF	R - H	ISSF	R - V	/SSF	L - V	SSF	L - H	SSF	R – H	SSF	R - V	SSF
Period	A	S	Α	S	A	S	A	S	A	S	A	S	A	S	A	S
1	63.1	16.8	67.0	21.7	64.0	25.6	59.3	16.5	58.9	13.0	61.0	23.2	58.0	25.8	55.3	15.9
2	59.9	13.2	66.0	2.0	78.0	6.5	68.3	13.5	52.7	12.4	61.0	1.5	68.0	3.8	59.5	12.4
3	76.5	16.2	82.0	19.7	82.0	4.8	92.1	9.9	68.1	30.7	79.0	23.9	78.0	7.2	86.1	16.5
4	79.0	21.4	87.0	2.9	93.0	5.0	78.0	19.7	75.6	19.7	87.0	1.2	91.0	2.0	74.8	20.9
5	87.0	13.0	96.0	4.8	96.0	4.5	83.9	10.9	84.6	10.3	95.0	3.9	96.0	3.5	83.8	8.7
9	86.7	12.4	99.0	0.5	98.0	1.7	90.9	10.1	86.5	8.4	98.0	1.0	97.0	1.6	88.6	7.6
				PE of	${ m fN_{tot}}$							PE of	$\mathbf{P}_{\mathrm{tot}}$			
	L – J	/SSF	L - F	ISSF	R - H	ISSF	R - V	/SSF	L - V	SSF	L - H	SSF	R – H	SSF	R - V	SSF
Period	A	\mathbf{v}	A	S	A	S	A	S	A	S	A	S	A	S	A	S
1	39.0	13.8	53.0	21.8	54.0	20.8	41.1	7.7	68.8	35.3	97.0	3.8	96.0	4.9	69.4	25.8
2	47.4	11.0	46.0	11.2	60.0	14.1	51.3	12.2	55.2	21.2	96.0	0.1	99.0	0.1	58.3	22.1
3	61.2	40.2	58.0	17.6	58.0	11.3	78.5	25.4	62.9	10.4	96.0	1.8	99.0	0.6	78.6	8.0
4	74.7	15.7	78.0	2.1	79.0	1.2	74.3	17.3	67.0	19.7	92.0	3.0	99.0	0.7	70.5	19.1
5	76.1	8.9	79.0	8.3	81.0	6.5	74.1	8.9	50.9	12.0	71.0	8.3	78.0	7.4	58.2	17.1
9	81.4	5.7	87.0	3.8	87.0	4.7	76.2	6.9	63.3	9.1	71.0	11.1	77.0	3.9	69.0	7.1

Table 3.10. Purification efficiency (PE) of organic matter (after BOD₇ and COD_{cr}), N_{tot} and P_{tot} in Rämsi FS from November 2005 to

d ur onnz	stious 1-	-0. A – a	verage	o – slanc	laru uev	lauon. I	ror expr	anauons		c. J.Y.						
				Mr of	BOD_7							Mr of (OD_{er}			
	L – 1	VSSF	L - F	ISSF	R – H	ISSF	R - V	/SSF	L – J	/SSF	L - F	ISSF	R – H	ISSF	R - V	SSF
Period	A	S	A	S	A	\mathbf{N}	A	S	A	\mathbf{N}	A	S	A	\mathbf{v}	A	S
1	44.4	20.1	12.8	3.8	9.8	4.2	37.2	11.7	77.8	20.1	22.9	10.0	18.3	4.2	61.4	20.4
2	30.7	6.4	9.4	2.3	8.7	2.4	25.1	4.2	49.5	10.7	15.9	3.7	14.8	3.7	38.2	7.5
3	29.0	15.8	8.2	4.4	8.0	4.2	25.1	15.9	48.4	37.0	14.3	10.6	14.0	10.3	41.8	39.7
4	23.8	4.7	9.1	2.5	8.5	2.1	19.8	6.2	35.5	13.8	14.1	6.6	13.9	5.7	33.0	11.5
5	43.8	11.2	13.6	3.2	13.1	3.1	41.9	11.3	71.1	41.8	22.9	11.5	22.4	11.8	67.8	41.9
9	17.9	4.0	5.3	1.0	5.3	1.1	18.0	4.1	25.6	3.7	7.8	0.8	7.8	0.8	26.1	3.3
				Mr of	${ m fN_{tot}}$							Mr of	$f P_{tot}$			
	L - J	VSSF	L - F	HSSF	R – H	ISSF	R - V	/SSF	L – J	/SSF	L - F	ISSF	R – H	ISSF	R - V	SSF
Period	A	S	A	S	A	S	A	S	A	S	Α	S	A	S	A	S
1	3.4	2.9	0.2	1.4	0.4	1.5	2.2	3.1	1.4	0.8	0.5	0.3	0.8	0.2	1.7	1.0
2	1.8	1.9	0.7	0.7	0.6	0.6	0.6	1.7	0.3	0.3	0.2	0.1	0.4	0.1	0.4	0.3
3	2.0	1.0	0.5	0.3	0.4	0.2	1.1	0.9	0.3	0.6	0.2	0.2	0.2	0.2	0.3	0.5
4	2.8	1.4	1.0	0.5	0.7	0.5	2.5	1.4	0.3	0.4	0.3	0.2	0.4	0.2	0.4	0.3
5	5.7	2.7	1.9	0.7	1.8	0.7	5.9	2.2	1.8	1.2	0.7	0.3	0.7	0.3	1.8	1.2
9	2.7	0.4	0.9	0.1	0.9	0.1	2.6	0.4	0.6	0.3	0.3	0.1	0.2	0.1	0.5	0.2

Table 3.11. Mass removal rate (Mr) of organic matter (after BOD₇ and COD_{cr}), N_{tot} and P_{tot} in Nõo FS from November 2005 to December 2006 in periods 1–6. A – average: S – standard deviation. For explanation see Table 3.9

zuuo in pi	eriods 1–c	A - aV	erage; >	- stand	ard devi	ation. F	or explai	nation se	e ladie.	<i>3.</i> 9.						
				Mr of]	BOD_7							Mr of C	OD_{cr}			
	L - V	'SSF	L - H	ISSF	R – H	SSF	R - V	'SSF	L - V	SSF	L - H	SSF	R – H	SSF	$R - V_{2}$	SSF
Period	A	S	Α	S	А	S	Α	S	Α	S	Α	S	A	S	A	S
1	139.3	6.69	59.1	30.9	60.6	33.8	131.0	65.4	165.8	67.8	68.4	31.5	71.2	38.1	155.8	71.2
2	34.2	9.0	15.0	1.1	17.7	2.3	38.9	9.5	42.5	9.3	19.6	0.9	22.0	0.7	48.0	10.4
3	29.4	20.4	12.6	7.8	12.7	8.5	35.4	20.7	37.5	26.1	17.3	10.5	17.1	11.9	47.5	27.3
4	59.1	22.9	25.9	5.7	27.9	6.8	58.4	17.4	84.5	19.4	39.0	3.0	40.6	2.7	83.7	19.2
5	50.6	14.2	22.3	5.5	22.4	5.7	48.8	14.2	76.1	18.4	34.6	7.0	34.7	7.3	76.1	18.4
9	40.3	13.1	18.5	4.3	18.3	4.0	42.3	12.6	62.6	8.0	28.4	2.5	28.1	2.1	64.2	8.7
				Mr of	ΓN_{tot}							Mr of	$\mathbf{P}_{\mathrm{tot}}$			
	L - V	'SSF	L - H	ISSF	R – H	SSF	R - V	'SSF	L - V	SSF	L - H	SSF	R – H	SSF	$R - V_{2}$	SSF
Period	Α	S	A	S	A	S	A	S	A	\mathbf{v}	A	S	A	\mathbf{v}	A	S
1	7.9	4.2	4.3	2.1	4.9	2.8	8.3	3.0	5.3	2.7	3.0	1.3	3.0	1.2	5.3	2.7
2	2.7	1.1	1.0	0.4	1.3	0.6	2.7	1.1	0.9	0.5	0.6	0.1	0.6	0.1	0.9	0.6
3	1.9	1.4	0.7	0.6	0.7	0.7	2.4	1.3	0.8	0.4	0.5	0.2	0.5	0.2	0.9	0.4
4	4.4	1.1	1.8	0.2	1.9	0.2	4.4	1.1	2.0	0.7	1.1	0.2	1.2	0.3	2.1	0.6
5	4.0	1.2	1.7	0.5	1.7	0.4	3.9	1.0	1.2	0.3	0.7	0.2	0.8	0.2	1.4	0.4
9	4.2	0.5	1.9	0.2	1.8	0.2	3.9	0.7	1.3	0.4	0.6	0.2	0.6	0.2	1.4	0.4

Table 3.12. Mass removal rate (Mr) of organic matter (after BOD₇ and COD_{er}), N_{tot} and P_{tot} in Rämsi FS from November 2005 to December 2006 in neriods 1–6 A – average: S – standard deviation. For evolumation case Table 3.0

Table 3.13. Average value of COD_{cr}/N_{tot} ratio in the Nõo FS left and right parallel (Laber *et al.*, 2003 – theoretical minimal value: 2.8; optimal value in field conditions: 6.0). For an explanation of periods, see chapter 2.2.

Period		Ratio of COD	O _{cr} /N _{tot} in left par	allel of Nõo FS	5
	Inflow to	Outflow from	Outflow from	Inflow with	Inflow with
	VSSF	VSSF	HSSF	re-circ.	re-circ.
				from VSSF	From HSSF
1	9.5	4.8	3.6	7.5	6.6
2	10.5	4.2	3.2	4.7	4.3
3	14.8	5.1	4.0	11.5	11.0
4	11.7	10.6	5.7	11.4	10.2
5	10.1	5.2	2.9	8.9	8.9
6	7.5	3.3	2.6	6.5	6.7
Period		Ratio of COD	_{cr} /N _{tot} in right pa	rallel of Nõo F	S
	Inflow to	Outflow from	Outflow from	Inflow with	Inflow with
	VSSF	VSSF	HSSF	re-circ.	re-circ.
				from VSSF	From HSSF
1	9.5	5.8	5.2	7.8	7.4
2	10.5	5.2	3.9	5.2	4.6
3	14.8	5.8	3.9	11.0	10.6
4	11.7	10.5	4.2	11.3	9.3
5	10.1	6.8	3.1	9.3	8.7
6	7.5	2.6	2.9	6.3	6.9

The differences in the purification efficiencies between the two parallel systems in Nõo could be caused by the relatively poor properties of CL (larger grain size, dusty and different solubility properties) compared to LWA (smaller grain size and therefore higher surface area) in terms of aeration and insulation, which enhance the effects of other factors such as hydraulic loading rate and temperature regime. The CL and LWA filter system probably retained P through precipitation and sedimentation reactions with Ca-rich particles, whereas the LWA system retained P in redox-potential dependent reactions of P with soil Fe and Al (Zaytsev *et al.*, 2007).

We were able to meet the Estonian standards only for BOD_7 and COD_{cr} during the end of the project (period 5 and 6). In periods 5 and 6 we also obtained sufficient results for N_{tot} purification efficiency. The Nõo FS did not met regulation rates for P_{tot}. Considering the aims of the wastewater treatment regulations set by the Helsinki Commission, we met the limits set for BOD in almost all periods. Regarding COD_{cr} , N_{tot} and P_{tot}, in most periods we obtained results that did not assure the required concentration (mg Γ^1), but they met the targets that were set for purification efficiency (%).

<u>Rämsi FS</u> – the outlet concentrations from both parallel systems significantly exceeded standard values in the first periods, due to the high loading of the systems with nutrients and organic matter (Table 3.2. and Table

3.4.). This affected the system's purification efficiency (Table 3.10.) and mass removal rate (Table 3.12.).

In terms of purification efficiency, the right-hand parallel was more stable than the left-hand system. Better overall purification efficiencies in water quality parameters were noted in the 2^{nd} , 4^{th} and 5^{th} periods. There were also higher re-circulation rates in the right-hand parallel (Table 2.4.).

Values for COD_{cr}/N_{tot} ratio are presented in Table 3.14. High COD values could also affect the purification efficiency of N_{tot} . As can be seen, the abovementioned ratio in HSSF filters and inflow filters is above the recommended ratio of 6.0 (Laber *et al.*, 2003). Nevertheless, the loading rate of effluents was too high to achieve satisfactory N_{tot} purification efficiency.

The Spearman Rank Order Correlation analysis shows a significantly positive correlation in both parallel FSs with re-circulation and BOD₇, COD, N_{tot} and NH_4 -N purification efficiency, which was expected. There was also a significant negative correlation with re-circulation and P_{tot} purification efficiency in both FSs.

The results show that higher re-circulation is a good solution to improve purification performance in overloaded systems. On the other hand, an overly high re-circulation rate (in the case of Rämsi, up to 600%) has a negative effect on P_{tot} removal. Inorganic chemical reactions such as phosphorus adsorption and precipitation are normally rapid processes that are not greatly affected by increasing wastewater-media contact time. Therefore the use of effluent recirculation may have little impact on P_{tot} removal (Sun *et al.*, 2003). As found in Rämsi, the effect of re-circulation can even turn in the opposite direction. Calcium present in the wastewater itself can promote phosphorus precipitation (Maurer *et al.*, 1999). This is probably the explanation for the very effective phosphorus removal in the pilot-scale system at Rämsi pig farm. Since the pig fodder also contains Ca-minerals (according to the pig farm data, average Ca content is 11.3 g kg⁻¹), it also contains phosphorus (average 8.6 g kg⁻¹), and some of the Ca is excreted with the slurry. The Ca inside the wastewater probably allows the phosphorus to precipitate as Ca-phosphate.

We did not succeed in meeting Estonian standards on demanded concentration (mg l^{-1}), but we did at least obtain some results that met the margins of purification efficiency. In the right bed of Rämsi FS we did obtain outflow parameters < 1.5 mg l^{-1} in period 2. The results were almost the same when we compared with the HELCOM aims of wastewater treatment regulations. The only difference was that in the left system of Rämsi FS, the result for the 6 periods was BOD < 40 mg l^{-1} .

Period	Ratio of COD _{cr} /N _{tot} in the left parallel of Rämsi FS				
	Inflow to	Outflow from	Outflow from	Inflow with	Inflow with
	VSSF	VSSF	HSSF	re-circ.	re-circ.
				from VSSF	from HSSF
1	13.9	9.4	11.7	12.2	13.2
2	15.3	13.8	11.1	14.8	13.8
3	18.0	14.8	9.6	17.1	15.2
4	18.8	18.1	10.7	18.7	17.3
5	17.1	11.0	4.0	15.9	14.8
6	14.0	10.2	2.7	13.4	12.9
Period	R	Ratio of COD _{cr} /N _{tot} in the right parallel of Rämsi FS			
	Inflow to	Outflow from	Outflow from	Inflow with	Inflow with
	VSSF	VSSF	HSSF	re-circ.	re-circ.
				from VSSF	from HSSF
1	13.9	10.5	12.7	12.6	13.5
2	15.3	12.7	12.1	14.5	14.4
3	18.0	11.7	9.6	16.9	15.6
4	18.8	18.4	8.1	18.7	16.9
5	17.1	10.7	4.1	15.8	15.0
6	14.0	6.7	3.3	12.6	12.7

Table 3.14. Average values of the COD_{cr}/N_{tot} ratio in the left and right parallels of Rämsi FS (Laber *et al.*, 2003 – theoretical minimal value: 2.8; optimal value in field conditions: 6.0). For an explanation of periods, see chapter 2.2.

3.3. General discussion

Subsurface flow wetlands for domestic wastewater purification are most commonly used in Estonia. About 30 subsurface flow wetlands have been installed in Estonia to date (Vohla *et al.*, 2005; Vohla, 2008), and for that reason our studies focused on subsurface flow CWs and FSs. Free water surface (FWS) constructed wetland systems, where vegetation has a more significant role, are also used here in Estonia and throughout the world for wastewater treatment (Maddison *et al.*, 2005; Bojcevska and Tonderski, 2007; Bastviken *et al.*, 2009), but our purpose was to examine the performance capabilities of subsurface flow wetlands.

Zimnoch *et al.* (2004) did an Artificial Neural Networks (ANN) prediction analysis for N_{tot} and P_{tot} output concentrations for the Kodijärve CW, which covered the period from January 1997 to February 2002. The results for nitrogen are presented in Figure 3.4, from which it can be concluded that total nitrogen concentration exerted the strongest influence on nitrogen concentration in the wetland outflow during that period. In contrast, the impact of parameters such as BOD, pH, Total Suspended Solids (TSS or SS) and flow rate (Q) was small. Analyzing different nitrogen components, the NH₄-N signal was shown to have a greater influence on nitrogen output concentration than the NO₂-N and NO₃-N signals. This result is consistent with other approaches (Noorvee *et al.*, 2007; Publication I), and can be explained by the insufficient aeration of the wastewater in the wetlands.



Figure 3.4. Contribution of input water parameters to total nitrogen concentration at the output from the Kodijärve wetland, by Zimnoch *et al.* (2004).

Zimnoch *et al.* (2004) also analyzed the residence time of N and P in wetlands. The total nitrogen signal at the output from the wetland consisted of two contributions. The first is rapid and represents a residence time of ca. 2-3 months. The other has a value of about 9-11 months and is stronger than the first. In the case of phosphorus, residence time in the wetland is longer than 12 months.

During the second period (K2), the HSSF filter material become saturated by phosphorus at depths (Vohla *et al.*, 2007) and phosphorus PE was reduced compared with the 1st period (K1). From 1997 to 2004 we collected soil samples from Kodijärve HSSF filters, from soil layers 0–10 cm, 30–40 cm and 60–70 cm (one procedure covered a total of 54 samples). The results are shown in Figure 3.5, and on the basis of these it is clear that especially phosphorus accumulation in the filter material was particularly high at the end of analysed 2^{nd} working period (October 2002 – April 2005).

Mander *et al.* (2008) pointed out that the Kodijärve HSSF CW is a strong C sink, with annual C sequestration of 649 and 484 kg C year⁻¹ per wetland in 2001 and 2002 respectively. About 1.5–2.2 kg C m⁻² was incorporated in the phytomass and/or soil of this wetland system. As C emissions are low in winter (Teiter and Mander, 2005), the mineralization of the previous year's litter occurs during the vegetation period, when CO₂ fluxes from decomposing litter and other sources are newly available for plant photosynthesis. The great difference in soil C accumulation in 2001 and 2002 is most probably affected by the high variability of plant NPP (Net Primary Production) due to changes in

environmental conditions. For instance, the warmer, sunnier and drier vegetation period in 2002 was the most probable reason why the Kodijärve HSSF CW showed significantly higher plant production and respective C sequestration that year. Lower plant NPP in 2001 led to a smaller litter amount as a source for C sequestration. During the cooler and less sunny vegetation period in 2001, the plant assimilation was about 2 times lower than the accumulation in soil and bacteria. This is, however, also influenced by variations of C inputs with wastewater.

In the 3rd period (K3), P binding was probably low because the LWA used in Kodijärve CW HSSF filter systems is produced from local clay mineral in Estonia (trademark name Filtralite S, M, L), and does not possess the characteristics of Filtralite-P (see also chapter 2.5.). The usable surface of the filter was also smaller than in previous periods (Table 2.2.).



Figure 3.5. Nitrogen, phosphorus and organic material accumulation in the soil of Kodijärve CW from 1997 to 2004 (working periods K1 and K2).

In VSSFs, re-circulation and especially the higher re-circulation rate of the wastewater significantly improves aeration and overall purification efficiency. The re-circulation rate must be at least 100 to 200 percent of the inflowing wastewater in order to achieve satisfactory results in terms of effective BOD₇ and COD_{cr} removal and nitrification. Those results which are achieved by implementation of re-circulation are fitting with results which are described by other authors in their published articles, see chapter 1. We observed a significant positive correlation between the re-circulation rate and purification between PE and water temperature was also noticeable, for example in the Nõo FS. However, the positive effect of water temperature on purification efficiency could also partly be explained by the fact that the application of higher re-circulation rates was in part concurrent with the rise in water temperatures

during the test period. Also, the filters must not be overloaded in terms of hydraulic and mass loading rates (Noorvee *et al.*, 2007; Publication I, Põldvere *et al.*, 2008, 2009, 2010; Publications II, III, IV).

Nitrification occurs in the filters after carbon is reduced, and therefore that is only possible if there is enough oxygen available in the system after COD (BOD) elimination (Laber et al., 1999). Insufficient oxygen content is considered to be one of the most important factors hampering nitrogen removal. The main reasons for unsatisfactory results in the case of both filter systems during the first periods (operational regimes) were the overloading (in terms of both hydraulic, nutrient and organic matter load) and/or low re-circulation rates applied (Põldvere et al., 2008, 2009, 2010; Publications II, III, IV). Likewise, the communities of microorganisms were insufficiently developed in the filter material. Cooper et al. (1999) also recommended pumping effluent back from the outlet of the VSSF to improve the denitrification. In terms of applying predenitrification, a better solution would be to pump the wastewater into the septic tank, where more organic matter is available for denitrification (Põldvere et al., 2009, Publication III). In order to achieve high removal of N_{tot}, re-circulation of treated wastewater back to the inlet of the septic tank is suggested (Johansen et al., 2002). The raw wastewater mixes with the nitrified water in the settling tank. The raw wastewater contains the necessary carbon source and anoxic conditions for denitrifying bacteria (Laber et al., 2003). Since the change in the recirculatioon pattern in the case of the hybrid filter system (interim + outflow well or only the outflow well) showed no significant effects on purification performance, the use of re-circulation from the interim well seems to be irrelevant in terms of the additional improvement of purification performance, and back-pumping only from the outflow well is more adequate (Põldvere et al., 2009, Publication III). Regarding filter material, Siim (2008) found that LWA is not as effective in absorbing $NH_{4}N$ as CL gravel, whereas gravel adsorption capability is higher per cell.

If one applies higher re-circulation rates, the hydraulic constraints of the filter material must also be taken into consideration. Typical hydraulic loading for VSSFs is 40–500 mm d⁻¹ (Kadlec *et al.*, 2000) or 100–400 mm d⁻¹ (Paing *et al.*, 2006). Average recommended hydraulic loading for HSSF CWs varies from 20 to 100 mm d⁻¹ (Kadlec *et al.*, 2000). During the study period no negative side-effects such as clogging were observed when the higher recirculation rates were applied.

Higher re-circulation rates did not significantly affect the phosphorus removal rate. Inorganic chemical reactions, such as phosphorus adsorption and precipitation, are normally rapid processes that are not greatly affected by the increasing of wastewater-media contact time (Noorvee, 2007). Therefore the use of effluent re-circulation may have little impact on P_{tot} removal (Sun *et al.*, 2003). Nevertheless, a very high re-circulation rate (up to 600%), when the filter material acts as a phosphorus precipitating substrate, has a negative effect on P_{tot} removal (Põldvere *et al.*, 2009 and 2010, Publication III and IV).

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Unfortunately, the LWA (Filtralite S, M, L) used as filter material rapidly lost its phosphorus adsorption and sedimentation properties. In LWAs, especially in Filtralite P, Ca-minerals are a very important additive for the precipitation of phosphorus. In contact with water, this may yield a dramatic increase in pH (pH up to 12), which may favour the precipitation of phosphates (Jenssen and Krogstad, 2003). We did not register great changes in pH in our systems. Thus for future prospects it is very important to find a suitable filter material for phosphorus removal via adsorption or precipitation. For that purpose we may follow the results which are acquired by Vohla (2008), who found good average P accumulation ability when testing with oil-shale ash and peat. Prochaska *et al.* (2006) also for example detected that sand and dolomite had good ability to accumulate phosphorus as follows 121 mg kg⁻¹ and 168 mg kg⁻¹.

Another possibility to assure sufficient phosphorus removal (<1.5 mg l⁻¹ in the outflow) is to use chemical precipitation inside the septic tank (Brix and Arias, 2005). As mentioned in chapter 2.1, the Rämsi FS was disassembled after December 2006. The Nõo FS was conserved until August 2007, when the last experiments began to test the chemical precipitation of phosphorus using aluminium sulphate [Al₂(SO₄)₃] in the septic tank (Kivisild, 2009). The average phosphorus concentrations in the inflow were 19.4 mg l⁻¹(loading rate 5.8 g d⁻¹). For chemical deposition a 7.9%-solution of aluminium sulphate of 321.4 ml d⁻¹ (33.2 g d⁻¹) was applied. The achieved PE for phosphorus removal was over 90% (1.0 to 1.3 mg l⁻¹; Kivisild, 2009). Nevertheless, it is still necessary to find a proper way to use septic tank mud.

The construction costs of such CWs and FSs filled with LWA (Filtralite S, M, L) would, in Estonia, range from 1279 to 1051 \in pe⁻¹ (for 50 to 100 pe). Compared to AWP with equivalent loading rates (for 50 to 100 pe) and treatment performance, consisting of concrete basins for nitrification, secondary sedimentation and sludge thickening, with construction costs of 1432 to 1112 \in pe⁻¹, CWs would be cheaper to establish. If we also consider operating costs, such as electrical power, chemical, labour, sludge treatment costs and depreciable cost, which total 84 to 72 \in y⁻¹ for CWs and 129 to 96 \in y⁻¹ for AWPs, then it is reasonable to select CWs for communities from 50 to 100 pe. This conclusion is based on a lifetime assessment of different treatment systems. In the evaluation of depreciable cost, the lifetime of facilities is considered to be 40 years, although in the case of CWs a lifetime of 20 years is assumed for the filter material (Põldvere *et al.*, 2009; Publication III).

4. CONCLUSIONS

We analysed the performance of one full-size VSSF and HSSF CW and two pilot-scale VSSF and HSSF FS systems in Estonia. We observed in detail the removal of organic material (after BOD_7 and COD_{cr}), nitrogen (N_{tot}) and phosphorus (P_{tot}) from wastewater (see publications I–IV). This thesis is mainly concentrated on the results that were achieved from November 2005 to December 2006 (divided into 6 periods of various re-circulation regimes), but for background a review of the long-term (January 2007 to May 2008, divided into three working periods) performance of Kodijärve CW is provided. A short overview of experiments performed in the Nõo FS from Aug 2007 to March 2008 is also included.

We can conclude that in cold climates it is important to use a VSSF system before the HSSF system, because better aeration of wastewater provides overall better purification efficiency (PE) considering BOD₇, COD_{cr} and N_{tot} removal. It is also necessary not to overload filters in terms of both hydraulic and mass loading rates, so CW and FS dimensions must match the characteristics of the effluent.

VSSF and HSSF filters can be designed in an economical way, especially when wastewater re-circulation of 100–300% is applied. The re-circulation of the wastewater improves overall purification significantly. It is possible to achieve satisfactory results in terms of effective BOD and COD removal and nitrification/denitrification as well as P_{tot} removal. We observed a significant positive correlation between the re-circulation rate and purification efficiency of BOD₇, COD_{cr}, N_{tot} and NH₄-N.

In the Nõo FS continuous flow hybrid CL and LWA filters for the treatment of municipal wastewater, the mean purification efficiencies (PE, %) of BOD₇, N_{tot} , P_{tot} during the test period from Nov 2005 to Dec 2006 were 82%, 47% and 66% respectively for CL and LWA FS, and 87%, 51% and 55% respectively for LWA FS. The highest purification efficiencies, 99% and 82% for BOD₇ and N_{tot} removal respectively, were achieved in the LWA FS, when the re-circulation rate of 300% was applied. In VSSFs, LWA was a better filter material than crushed CaCO₃-rich limestone, because of its higher surface area. Better aeration and insulation enhance the effects of other factors, such as hydraulic loading rate and temperature regime.

In the Rämsi FS the highest purification efficiencies, 98.5% and 87% for BOD₇ and N_{tot} removal respectively, were achieved in both Rämsi FSs, when the re-circulation rate of 300 to 600% was applied.

In the Kodijärve CW the highest purification efficiencies, 98% for BOD₇ removal, were achieved when the re-circulation rate of 300% was applied and 58% for N_{tot} removal were achieved when the re-circulation rate of 150% was applied. Considering Kodijärve CW performance from January 1997 to May 2008, best PE results were achieved in the 2nd period (K2), when PE of BOD₇ was 97.3% and PE of N_{tot} was 53.2%. In the 3rd period the results were not so

good, because the CW surface area was smaller and mass loading rates were therefore higher.

The application of re-circulation makes it possible to avoid overdimensioning CWs, which leads to the establishment of more cost-effective CWs and FSs. Our calculations show that a CW wastewater treatment system for at least 50 to 100 person equivalents would be more cost effective to establish and maintain than an active sludge wastewater treatment plant.

The main factors affecting the performance of the continuous flow hybrid CW and FS as concerns BOD_7 and N_{tot} removal were water temperature and recirculation rate, in which the effect of recirculation and thus retention time was more noticeable. There was no clear influence of other influencing factors such as water temperature, hydraulic and mass loading rate on purification efficiency during the test period, as the filter systems were tested in field conditions using real wastewater. Also no significant seasonal effects on purification efficiency of the water quality indicators were found. Thus the mass loading rate of filter systems was determined on the basis of the characteristics of wastewater and applied hydraulic loading rate. For future design suggestion in terms of applying pre-denitrification, a better solution would be to pump the wastewater into the septic tank, where more organic matter is available for denitrification.

Our study provides forward guidelines to develop systems that match Estonian legislation, when treated wastewater is channelled into a body of water (the subsequent reuse of treated wastewater is not common in Estonia). With pilot systems we did not sufficiently achieve the goals set by the Estonian and Helcom regulations regarding N_{tot} and P_{tot} removal from wastewater, but it is possible that in future developments the results of these experiments will be followed.

Since the phosphorus retention capacity of the Kodijärve HSSF CW reached its limit, the filter material of the HSSF was replaced, using light-weight aggregates (LWA) in July 2005. Once the media is saturated with P, the entire system would need to be removed and replaced in order to restore P removal performance. The LWA used as filter material in CWs and FSs did not offer better results in phosphorus removal. In LWAs, Ca-minerals are a very important additive for the precipitation of phosphorus. In contact with water, this may yield a dramatic increase in pH (pH up to 12), which may favour the precipitation of phosphates. We did not register great changes in pH in our systems. For sufficient P_{tot} removal, it is crucial that the proper filter material for P removal via adsorption or precipitation be used. Another possibility is to use chemical precipitation inside the septic tank, which turned out to be an effective way. During experiments from August 2007 to March 2008 in Nõo FS, high PE values for P (>90% (1.0 to 1.3 mg P Γ^{-1} in the outflow) were achieved.

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SUMMARY IN ESTONIAN

Orgaaniliste ainete, lämmastiku ja fosfori ärastamine reoveest hübriidsetes pinnasfiltersüsteemides

Euroopas on tehismärgalasüsteeme, mis puhastaksid erinevat tüüpi reovett, uuritud juba 1950-ndatest aastatest. Käesolevas doktoritöös on analüüsitud ühe täismõõdulise vertikaalvoolulise ja horisontaalvoolulise hübriidse pinnasfiltersüsteemi (Kodijärve) ning kahe vertikaalvoolulise ja horisontaalvoolulise hübriidse piloot- ehk katseseadme (Nõo ja Rämsi) funktsioneerimist reoveepuhastuse eesmärkidel.

Detailsemalt analüüsiti orgaaniliste ainete (BHT₇ ja KHT_{cr} alusel), lämmastiku (N_{üld}) ja fosfori (P_{üld}) ärastamist reoveest (artiklid I–IV). Peamiselt kontsentreeruti andmetele, mis koguti perioodil november 2005 – detsember 2006 (periood jaotati 6 alamperioodiks). Taustateabena anti ülevaade pikaajalistest (jaanuar 1997 – mai 2008) tulemustest, mis on kogutud Kodijärve pinnasfiltersüsteemist ja jaotatud kolme perioodi. Samuti esitatakse ülevaade eksperimentidest, mis viidi läbi Nõo katseseadmel perioodil august 2007 – märts 2008.

Doktoritöö põhjal võime järeldada, et oluline on kasutada vertikaalvoolulist pinnasfiltrit enne horisontaalvoolulist pinnasfiltrit, kuna parem reovee hapnikuga varustatus tagab paremad tulemused puhastusefektiivuste osas, eriti BHT₇, KHT_{cr} ja N_{üld} näitel. Oluline on filtersüsteeme mitte üle koormata ja seda nii hüdrauliliselt kui ka reoainete koormuse osas ehk pinnasfiltersüsteemide dimensioonid peavad vastama neisse juhitava reovee parameetritele.

Vertikaal- ja horisontaalvoolulisi pinnasfiltersüsteeme saab rajada ökonoomselt, eriti juhul kui rakendatakse reovee tagasipumpamist vähemalt määral 100–300% filtritesse juhitud reovee hulgast. Reovee tagasipumpamine parandab üldist puhastusmäära. Võimalik on saavutada rahuldavaid tulemusi BHT ja KHT ärastamise ning nitrifikatsiooni ja denitrifikatsiooni osas.

Nõo katseseadmes, mis puhastas olmereovett, olid katseperioodil novembrist 2005.a. detsembrini 2006.a. keskmised puhastusefektiivsused BHT₇, N_{üld} ja P_{üld} osas järgnevad: 82%, 47% ja 66% vasakus paralleelsüsteemis ning 87%, 51% ja 55% paremas paralleelsüsteemis. Vasak paralleelsüsteem koosnes lubjakivikillustikuga täidetud vertikaalvoolulisest filtrist ja Eestis toodetud kergkruusaga täidetud horisontaalvoolulisest filtrist, kusjuures parempoolses süsteemis olid mõlemad filtrid täidetud Eestis toodetud kergkruusaga. Parimad puhastusefektiivsuse näitajad (BHT₇ 99% ja N_{üld} 82%) saavutati, kui parempoolses filtersüsteemis rakendati reovee tagasipumpamise määra 300%. Kasutatud kergkruus oli lubjakivikillustikust konkreetsel juhul parem materjal, kuna omas suuremat eripinda ning paremaid aeratsiooni- ja isoleerimisomadusi. Seetõttu mõjutati ka teisi parameetreid nagu vee voolavust filtris ja selle temperatuuri režiimi. Rämis katseseadmes saavutati BHT₇ ja N_{uld} osas 98,5% ja 87% puhastusefektiivsus mõlemas paralleelsüsteemis, kui rakendati reovee tagasipumpamisemäära 300 – 600%.

Kodijärve pinnasfiltersüsteemis saadi parimad tulemused BHT₇ puhastusefektiivsuse osas (98%), kui rakendati reovee tagasipumpamise režiimi 300% ja N_{üld} puhul 58%, kui reovee tagasipumpamise määr oli 150%. Pikaajalise (Jaanuar 1997. a. – Mai 2008. a.) tööefektiivsuse analüüsi põhjal osutus parimaks teine periood, millal BHT₇ puhastusefektiivsus oli 97,3% ja N_{üld} 53,2%. Kolmanda pikaajalise perioodi tulemused ei olnud nii head, kuna pinnasfiltersüsteemi pindala oli väiksem ja seetõttu oli reoainete koormus filtersüsteemile suurem.

Võib väita, et reovee tagasipumpamise rakendamine võimaldab mitte üle dimensioneerida pinnasfiltersüsteeme, mis loob eeldused ka majanduslikult tasuvate pinnasfiltersüsteemide rajamiseks. Tehtud tööde käigus läbi viidud kalkulatsioonid näitavad, et ratsionaalne on eelistada pinnasfiltersüsteeme aktiivmudapuhastitele kindlasti juhul kui puhastatava reovee reostuskoormuse pärineb asumitest 50–100 inimekvivalenti (ie). Toodud juhtudel on sellised süsteemid säästlikumad nii ehituslikke aspekte kui ka hooldust arvestades.

Teostatud uurimistöö alusel on peamisteks faktoriteks, mis mõjutavad BHT₇ ja N_{uld} ärastust, vee temperatuur, tagasipumpamise määr ja selle kaudu ka viibeaeg. Statistiliselt olulist seost ei leitud aga välisõhu temperatuuri ja puhastusefektiivsuse vahel. Tulevaste süsteemide puhul tuleks kaaluda reovee tagasipumpamist otse septikusse, kus on sobivaimad tingimused (piisavalt orgaanikat ning vahelduvad anaeroobsed ja anoksilised tingimused) efektiivseima denitrifikatsiooni kulgemiseks.

Pilootseadmetega teostatud katsete käigus ei saavutanud me täielikult tulemusi, mis vastaksid N_{uld} ja P_{uld} osas kehtivatele või tulevikus soovitatavatele HELCOMi nõuetele. Siiski pakuvada saadudud andmed edasiste uuringute ja arenduste jaoks kaalukat tuge.

Katsetel, mis algasid 2005. a. novembri kuus, kasutatud kergkruus ei omanud kõige efektiivsemat fosfori sidumise võimet. Piisava P_{uld} ärastuse tagamiseks on oluline valida selleks sobivad filtermaterjalid, et tagada efektiivset adsorptsiooni ja sadestumist. Üheks võimaluseks on ka sadestamine fosfori sadestamine septikusse kemikaali kasutamise teel. Viimane variant osutus efektiivseks alternatiiviks, mida tõestati katsete käigus, mis toimusid 2007. a augustist kuni 2008 märtsini Nõo filtersüsteemis. Süsteemist väljavoolavas heitvees saavutati $P_{uld} >90\%$ vähenemine ning kontsentratsioonid 1,0–1,3 mg/l.

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Publications

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DISSERTATIONES TECHNOLOGIAE CIRCUMIECTORUM UNIVERSITATIS TARTUENSIS

- 1. Sille Teiter. Emission rates of N₂O, N₂, CH₄ and CO₂ in riparian grey alder forests and subsurface flow constructed wetlands. Tartu, 2005.
- 2. **Kaspar Nurk.** Relationships between microbial characteristics and environmental conditions in a horizontal subsurface flow constructed wetland for wastewater treatment. Tartu, 2005.
- 3. **Märt Öövel.** Performance of wastewater treatment wetlands in Estonia. Tartu, 2005.
- 4. Alar Noorvee. The applicability of hybrid subsurface flow constructed wetland systems with re-circulation for wastewater treatment in cold climates. Tartu, 2005.
- 5. Christina Vohla. Phosphorus removal by various filter materials in subsurface flow constructed wetlands. Tartu, 2008.
- 6. **Martin Maddison.** Dynamics of phytomass production and nutrient standing stock of cattail and its use for environment-friendly construction. Tartu, 2008.
- 7. Marika Truu. Impact of land use on microbial communities in Estonian soils. Tartu, 2008.