Performance and Sustainability of Short-Rotation Energy Crops Treated with Municipal and Industrial Residues

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Abstract

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The sustainability of short-rotation willow coppice (SRWC) as a multifunctional system for phytoremediation—the use of plants for treatment of contaminated air, soil or water—and for producing energy biomass, was studied. SRWC is grown commercially in Sweden to produce energy biomass, nutrient-rich residues being applied as cost-efficient fertiliser to increase production. The principal residues used are municipal wastewater, landfill leachate, industrial wastewater (*e.g.* log-yard runoff), sewage sludge and wood-ash. Small- and large-scale experiments with residues aimed to quantify the extent of potential hazards and the performance of SRWC in reducing them.

Lysimeter experiments with willow plants, intensively irrigated with N-rich municipal wastewater, showed that N-leaching is a potential threat when high N loads are applied. Experimental data from SRWC fields irrigated with municipal wastewater in central Sweden suggest that in practice, N-leaching is significantly lower, even when the N load applied is greater than the N requirements of SRWC. Growth of willow plants of five different clones in pot experiments irrigated with landfill leachate was reduced by comparison with that of control plants. The reduction was attributed to saline stress or P deficiency, and indicates that, when hazardous compounds are present in wastewater, irrigation rates should be adjusted to avoid growth reduction. Genetic differences were observed between willow clones in salt tolerance and growth performance. The careful selection of clones to suit specific situations is therefore recommended. Leaf length can be used for rapid diagnosis of stress, to permit adjustment of the irrigation rate, and thus to avoid growth reduction. Phytoremedial efficiency of SRWC is satisfactory when the concentration of hazardous compounds in wastewaters is low, as in log-yard runoff, and depends on irrigation intensity. Application of sludge–ash mixtures to SRWC is not a substantial threat to sustainability, in terms of heavy metals. Total Cd in the soil is expected to decrease after harvest, but other metals and P loads, must also be considered.

Any decision concerning residue application must take into account factors such as residue composition, soil type, climate, and species or clone characteristics, if both a high growth rate of SRWC and sustainability are to be attained.

Keywords: Ash, heavy metals, landfill leachate, log-yard runoff, phytoremediation, *Salix*, short-rotation coppice, sludge, wastewater, willow.

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To my family

Contents

Appendix

Papers I-V

The present thesis is based on the following papers, which will be referred to by their Roman numerals:

I. Dimitriou, I. & Aronsson, P. 2004. Nitrogen leaching from Short-Rotation Willow Coppice after intensive irrigation with wastewater. *Biomass and Bioenergy*, 26 (5), 433-441.

II. Dimitriou, I., Aronsson, P. & Weih, M. Stress tolerance of five willow clones after irrigation with different amounts of landfill leachate. *Bioresource Technology* (In Press).

III. Dimitriou, I., Eriksson, J., Adler, A., Aronsson, P. & Verwijst, T. Fate of heavy metals after application of sewage sludge and wood-ash mixtures to short-rotation willow coppice (submitted to *Environmental Pollution*).

IV. Jonsson, M., Dimitriou, I., Aronsson, P. & Elowson, T. 2004. Effects of soil type, irrigation volume and plant species on treatment of log yard run-off in lysimeters. *Water Research* 38 (16), 3473-3644.

V. Jonsson, M., Dimitriou, I., Aronsson, P. & Elowson, T. Treatment of log yard run-off by irrigation of grass and willows. *Environmental Pollution* (In press).

Introduction

Short-rotation willow coppice in Sweden

Short-rotation willow coppice (SRWC) is grown in Sweden to produce biomass for energy. The crop is commercially grown on approximately15,000 ha of agricultural land, and the biomass produced is used in district heating plants for combined heat and power production. Cultivation of SRWC is fully mechanised from planting to harvest. In the initial phase, *ca.* 15,000 cuttings ha^{-1} are planted in double rows (distance between and within rows 1.5 m and 0.75 m, respectively, distance between cuttings 0.6 m), to accommodate future weeding, fertilisation and harvesting. Conventional inorganic fertilisers are commonly applied in the years following planting. Harvesting, by means of specially designed machines, takes place every 3–5 years, during winter when the soil is frozen. The aboveground biomass is chipped on-site, then stored or directly transported to the power plants. After harvest, the plants resprout vigorously, and replanting is therefore not necessary. The estimated economic lifespan of a SRWC is 20–25 years (Sennerby-Forsse & Johansson, 1989; Danfors, Ledin & Rosenqvist, 1998). In Sweden, different clones and hybrids of *Salix viminalis* L., *S. dasyclados* Wimm. and *S. schwerinii* E.L. Wolf. (Agrobränsle, 2005; SCB, 2005) are mainly cultivated. On commercial sites, biomass production is *ca.* 6–10 t ha^{-1} yr^{-1}, depending strongly on site conditions (Willebrand, Ledin & Verwijst, 1993).

Research conducted in Sweden after the oil crisis in the 1970s, with the aim of replacing fossil fuels by new energy sources, resulted in the introduction and cultivation of SRWC. The intention was to find fast-growing species which could be grown intensively and which would attain high growth rates. Extensive research into the plant biology, stand ecology and production systems of species from different genera (*Alnus*, *Betula*, *Populus*, *Salix, etc.*) suggested that willows grown in coppice performed better than other species tested, and were more suitable for use in Swedish energy forestry (Sirén *et al.*, 1987; Sennerby-Forsse, 1994). Nutrient utilisation and crop management were more efficient for willow plantations than for other species, and SRWC proved to be a sustainable way of producing CO_2-neutral fuels (Ericsson, 1994; Christersson, 2004).

SRWC as a multifunctional phytoremediation system

The use of wastewater for irrigation of crops is not a novel approach; advanced hydraulic systems for the collection and subsequent application of wastewater to farmland can be traced to the Mycenaean and Minoan civilisations in ancient Greece (Angelakis, Koutsoyiannis & Tchobanoglous, 2005). The transport and use of wastewater for irrigating agricultural crops was also recorded during the Middle Ages (Alker, 1999). In recent years, research programmes concerning the irrigation of large-scale tree plantations with wastewater were developed in Poland from the 1960s onwards (Kutera & Soroko, 1994). In Sweden, to achieve a more cost-efficient cultivation of SRWC for intensive production of energy biomass, it was necessary to reduce the input of conventional fertilisers. The use of nutrient-rich residues as an alternative, cost-efficient fertilisation method was proposed from the early stages of the development of SRWC, and has been successfully practised (Perttu, 1992; Aronsson & Perttu, 2001; Hasselgren, 2003). The principal residues used in Sweden are municipal wastewater, landfill leachate, industrial wastewater (*e.g.* log-yard runoff), sewage sludge and wood-ash. The benefits of using SRWC for phytoremediation, *i.e.* the use of plants for treatment of contaminated air, soil or water (EPA, 2000; Licht & Isebrands, 2005), are both environmental and economic. The residues, which are considered more as resources than as wastes, are dealt with more cost-efficiently as compared with conventional treatments, and the nutrients contained in the residues are used as low-cost fertilisers to increase plant production. The basic idea is to reduce the content of pollutants, nutrients or both in waters and soils by plant uptake and by filtration, and to facilitate microbial transformation. When used for phytoremediation, SRWC offers advantages such as high biomass yields and frequent harvests, which result in a substantial removal of hazardous compounds in the harvested plant biomass. The high evapotranspiration rate of SRWC (Persson & Lindroth, 1994) and the tolerance of their root systems to anaerobic conditions (Jackson & Attwood, 1996), make it possible to apply high irrigation rates. In addition, SRWC is reported to be capable of taking up substantial amounts of heavy metals, such as Cd—and is therefore suitable for remediating polluted sites (Landberg & Greger, 1996; Klang-Westin & Eriksson, 2003)—and is also capable of retaining large amounts of nutrients (Aronsson, 2000).

Sustainability of SRWC phytoremediation systems

The principle of sustainable development was first stated by the World Commission on Environment and Development, in a report (often called the 'Brundtland report') in 1987, as 'the development that meets the needs of present without compromising the needs of future generations'. This general definition was expressed in political terms and has been further developed to cover different aspects of sustainable practices. Sustainable use implies the 'use of the environment and its living resources at a rate that does not exceed its capacity for renewal in order to ensure its availability for future generations' (EIONET, 2005). According to EEA (2005), a broader understanding of sustainability in agriculture extends to the protection of landscapes, habitats, and biodiversity, and to overall objectives such as the quality of drinking water and air. Therefore, a sustainable application of residues to SRWC must ensure minimal environmental hazards to the plantations and the surroundings, not only for this generation but for future generations as well.

The indiscriminate application of certain residues to SRWC might result, for instance, in nutrient loads which would differ greatly from the nutrient requirements of the SRWC, and which in consequence would result in the accumulation of elements in the soil, in leaching to the groundwater or in losses to the air. Furthermore, hazardous compounds applied together with the residues may negatively affect the growth of SRWC, and pollute the surroundings. For example, the application of municipal wastewater—often regarded as one of the less hazardous residues applied to SRWC, since its nutrient content matches the requirements of willow (Aronsson & Perttu, 2001)—can result in a range of environmental problems. Examples of these are extensive nitrogen (N) leaching, which reduces the quality of drinking water and causes eutrophication of adjacent water systems; toxic effects of N on the plants, which cause biomass losses; or losses of N to the atmosphere, as nitrous oxide (N_2O) and ammonia (NH_3), which contribute to the 'greenhouse effect' and the eutrophication of water systems (Nielsen 1994; Mortensen, Nielsen & Jorgensen 1998; Börjesson, 1999). As regards landfill leachate, its usually high ionic strength may cause toxic effects on SRWC plants, while its high concentration of sodium chloride (NaCl) and other salts may lead to soil deterioration (Nixon *et al.*, 2001; Bowman, Clune & Sutton, 2002). The application of sludge–ash mixtures to SRWC may serve to increase heavy-metal and phosphorus (P) concentrations in the soil, despite the expected benefits to plant growth and to the physical characteristics of the soil (Labrecque, Teodorescu & Daigle, 1995; Shober & Sims, 2003). Industrial wastewater, such as log-yard runoff, has a relatively low hazard rating, with very low N concentrations, but contains organic compounds and P, which if leached to the groundwater and ultimately to surrounding water systems, may cause their quality to deteriorate (Borgå, Elowson & Liukko, 1996; Jonsson, 2004).

The special features of SRWC in phytoremediation systems, mentioned above, can in many cases reduce these risks. To achieve or improve the sustainable operation of SRWC phytoremediation systems, and simultaneously produce large quantities of biomass, an integrated approach should be adopted, which takes into account all of the contributing factors: the plants, the soil, the residue, the surroundings and the interactions between them.

Aims

The aim of this thesis is to investigate the performance and sustainability of SRWC as a multifunctional system for treatment of different types of residue (Table 1) and for the production of biomass for energy. Taking large-scale, multifunctional phytoremediation SRWC plantations as a guide, a series of small-scale experiments with different residues was conducted, to quantify the extent of potential hazards and the performance of SRWC in reducing them. Additional measurements in the field were made to test the applicability of the conclusions drawn from the experiments, and to identify improvements to the sustainability of the systems. An additional aim of this study was to show both the potential and the restrictions to the use of SRWC as multifunctional phytoremediation systems under a range of conditions, *e.g.* of application rate, type of residue, soil, climate, and species/clone, based on my own results and on results from literature related to my research.

The thesis is organised as follows: the account of each experiment, corresponding to an appendix paper, is prefaced with a short introduction to the relevant literature in the field. This is followed by a discussion of the results in relation to the findings of other authors. Finally, brief conclusions are presented.

Table 1. *Comparison of the relative (N=100) and of the actual concentrations of the various residues used in the experiments in Papers I-V. For wastewater from sludge dewatering (Paper I, Enköping), the values are averages from 1999, for landfill leachate (Paper II, Högbytorp), the values are averages from 2001, for log-yard runoff (Paper IV and V) values are averages between the two runoffs used in the two papers; and for sewage sludge and wood-ash (Paper III, Enköping), the values are averages for 2001*

Element	Optimum composition of *Salix**	Wastewater from sludge dewatering		Landfill leachate		Log-yard runoff		Sewage sludge		Wood-ash	
	%	%	mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%	mg g^{-1} DM	%	mg g^{-1} DM
N	100	100	925	100	270	100	1.5	100	29	100	0.1
P	14	7	61	1.5	4	173	2.6	117	34	17000	17
K	72	13	117	103	279	1466	22	4	1.2	32000	32

* Ericsson (1981).

Materials and Methods

In the following section, the materials and methods and the aims of the studies in the appended papers are briefly described. For details, the reader is referred to the appropriate section of the papers.

Nitrogen-rich municipal wastewater: a lysimeter study (Paper I)

Willow plants (*Salix viminalis*, clone '78-183') were planted in eight 1200-l lysimeters, four filled with clay (set A) and four with sand (set B). Two of the lysimeters from set A and two from set B were irrigated with N-rich wastewater, obtained by dewatering sewage sludge; the others were irrigated with the corresponding N doses of liquid fertiliser. Both irrigation waters had the same N concentration (*ca.* 400 mg l^{-1}), corresponding to 320 kg N ha^{-1}, applied during eight days. Thereafter, plants were irrigated daily for 123 days with tapwater at a rate of 10 mm d^{-1}. Drainage-water samples from each lysimeter were collected and analysed for nitrate-N (NO_3-N), ammonium-N (NH_4-N), organic N and organic C. Plant height increments were also measured.

The specific aims of this study were to quantify N-leaching following intensive application of wastewater under simulated 'worst-case' conditions, and to evaluate the risk of damaging the willow plants.

Landfill leachate: a pot-trial (Paper II)

Two cuttings of each of five willow clones ('78-183', 'Jorr', 'Loden', 'Olof', 'Tora') were planted in 1-dm^3 pots filled with sand, and irrigated with three different landfill leachate mixtures, diluted in different ratios and corresponding to 240, 180, and 120 mg Cl l^{-1}. For the control, liquid fertiliser (Blomstra, Cederroth International AB, Upplands Väsby, Sweden) was used, diluted to a concentration of 51 mg N-tot l^{-1}. The experiment was replicated in five blocks. All plants were supplied daily with 200 ml of the appropriate solution for approximately ten weeks. Additional tapwater was supplied to the plants to prevent water-stress. At harvest, each plant was separated into leaves, shoots, cutting and roots, then dried and weighed. Growth parameters, as well as leaf area, leaf length and leaf fluctuating asymmetry (non-directional deviation from expected leaf symmetry), were measured. The macronutrient content of plant parts was analysed for every clone, for the strongest leachate treatment and for the control.

The specific aims of this study were to quantify the growth response of the willow clones to irrigation with the various leachate solutions, and to discover a practical tool for identifying plant stress in the field.

Sewage sludge and wood-ash: field trials (Paper III)

Two commercial willow fields, planted with *Salix viminalis* (clone '78-021'), and in the first year of their second cutting cycle, were used. Both fields were situated in central Sweden, one at Lundby (16°55.98'E 59°39.40'N) and the other at Linnés Hammarby (17°46.77'E 59°49.44'N), on soils with a high clay content. The experimental treatments comprised two different amounts of a sludge–ash mixture, sludge only and ash only, plus a control. The sludge was dewatered, stabilised, municipal sludge and the ash was bottom-ash from a district heating plant where only biofuel is used. The experiments on both fields were identical, and consisted of 9×9 m plots in four blocks with four replicates of the same treatments. Soil, shoot and leaf-litter samples were collected at random from every plot annually and analysed for heavy metals (Cd, Cr, Cu, Ni, Pb, Zn). Biomass measurements were also made annually for every plot in both fields.

The specific aims of this study were to discover how heavy-metal uptake in SWRC was affected by the application of the sludge–ash mixture; and to investigate how the remediation effect of willow—a combination of growth rate and heavy-metal uptake—was affected by the various sludge–ash fertilisation treatments.

Log-yard runoff from a closed system: a lysimeter study (Paper IV)

Sixteen 1200-l lysimeters, eight with clay and eight with sand, were irrigated at two intensities (10 and 20 mm d^{-1}) with log-yard runoff from a closed log-yard sprinkling system, during approximately four months in the summer. Eight of the lysimeters were planted with willow (*Salix schwerinii* × *Salix viminalis*, clone 'Tora'), the other eight with common alder (*Alnus glutinosa* (L.) Gaertn.), resulting in eight pairs of lysimeters with the same combination of plant, soil and irrigation. Drainage water from every lysimeter was collected and analysed for total organic carbon (TOC), phenols, P and N. Retention in the lysimeters, absolute in g m^{-2} and relative in per cent, was calculated on the basis of concentration and volume differences between the irrigation water and the drainage water.

The specific aim of this study was to investigate possible differences in phytoremedial efficiency between willow and alder, and to determine any differences between clay and sand when irrigated with two intensities of log-yard runoff.

Log-yard runoff from an open system: lysimeter and field study (Paper V)

The field study was conducted in a field adjacent to the log-yard naturally grown with couch grass (*Elymus repens* L.), which was intensively irrigated with log-yard runoff. The irrigation rate was at most 66 mm d^{-1} during summer. Four pairs of groundwater tubes were installed to a depth of 1.2–3.1 m, inside and outside the irrigated couch grass area, for collection of groundwater samples. The same water as that applied to the field, was also applied to eight 68-l lysimeters, all of which contained soil columns collected from the field by means of a mechanical soil auger. Four of the lysimeters were planted, two with couch grass and two with willow (*Salix schwerinii* × *Salix viminalis*, clone 'Tora'), and were irrigated with the log-yard runoff at a rate of 49 mm d^{-1}. As control, three lysimeters of the same type grown with couch grass were irrigated with tapwater. Groundwater samples from the field, and drainage water from the lysimeters, were analysed for TOC, phenols, P and N. Retention in the lysimeters was calculated as in Paper IV, and retention in the field was estimated from irrigation and groundwater data, on the assumption of negligible transpiration, and uniform mixing in the groundwater.

The specific aim of Paper V was to investigate the phytoremedial efficiency of couch grass and willow when irrigated with log-yard runoff from an open sprinkler system.

Results and Discussion

Irrigation of SRWC with municipal wastewater

The direct discharge of treated wastewater to rivers and coastal waters is thought to be partly responsible for the eutrophication of water systems in Sweden, in consequence of the N and P content of the wastewater (SNV, 1997). It has been estimated that 21% of the total anthropogenic N load discharged into Swedish coastal waters is derived from wastewater treatment plants (SNV, 2003). The concentration of N, P and K in domestic wastewater was reported to match the optimal concentrations for *Salix* growth (Ericsson, 1981; Perttu & Kowalik, 1997); irrigation of SRWC with wastewater was considered an appropriate complementary method of reducing the total N amount discharged into water systems in Sweden. This would contribute to fulfilling the government's ambition to reduce N discharge into the sea by 30% of the 1995 level by the year 2010 (SNV, 2003). At the same time, SRWC cultivation produces biomass for energy, which also complies with proposals for the future energy supply in Sweden and the EU (SNV, 1998; EC, 2000).

The wastewater used in the experiment of Paper I was N-rich water produced by the dewatering of sludge in the wastewater treatment plant at Enköping, central Sweden (Table 1). This water, partly mixed with treated wastewater, is then used for drip irrigation of a 75-ha SRWC plantation adjacent to the wastewater treatment plant. The high N concentration in the wastewater makes storage during winter more cost-efficient owing to the smaller wastewater volume stored. Any assessment of the success and sustainability of wastewater irrigation on SRWC must take into account minimal leaching of N to the groundwater, a high growth rate of the SRWC, and no deterioration in soil and air quality. The results from the experiment were intended to provide information about the fate of N under 'worst-case conditions', *i.e.* when a high irrigation rate of N-rich wastewater, hence a high N load, is applied to SRWC during a short period.

Nitrogen leaching from willows grown in lysimeters (Paper I)

Nitrogen was leached from the lysimeters described in Paper I mainly in the form of NO_3^-, despite the fact that NH_4^+ was almost the only N-form present in the wastewater. This presumably depends on rapid nitrification to NO_3^-. The leaching of N from clay lysimeters corresponded to *ca.* 25% (*ca.* 80 kg ha^{-1}) of the applied N, irrespective of water type. The figure for the sand lysimeters was slightly higher. The performance of SRWC in terms of N-leaching should be compared with that of arable crops, the alternative crop on agricultural land, and with that of forests, since they compete as a producer of energy biomass. The mean fertilisation rate for arable crops in Sweden is *ca.* 120 kg of N yr^{-1} (Jordbruksverket, 2004) and the average leaching load of N reaches 22 kg N ha^{-1} yr^{-1}, which corresponds to *ca.* 20% of the applied N (Johnsson & Hoffmann, 1998). However, the concentration of NO_3^- in drainage water was far higher in the experiment of Paper I than the maximum concentrations observed from agricultural land (Hoffmann *et al.*, 2000). This can be attributed to the high irrigation load (10 mm d^{-1}) applied in the experiment of Paper I, which increases leaching, and the high N load (320 kg N ha^{-1}) described in Paper I. Generally speaking, a great advantage of SRWC in terms of N-leaching, as compared to arable crops, is the absence of tillage, which is responsible for the high level of N-leaching from most arable crops (Ledin, 1998; Joelsson & Kyllmar, 2002). Leaching of N from clearfelled forests in Sweden, which might be compared with harvested willow stands, is in the range <5–35 kg ha^{-1} yr^{-1} during one to five years after clearfelling, depending on the geographical situation, and is correlated with N deposition, which is in the range 10–30 kg ha^{-1} yr^{-1} (Akselsson, Westling & Örlander, 2004). Leaching from an established forest is considerably less (SNV, 2004), but this also applies to SRWC, notwithstanding the high N loads supplied by fertilisation (Aronsson & Bergström, 2001).

No significant differences were observed between wastewater and liquid fertiliser as regards leaching (Paper I). The only difference in the behaviour of the two water types was that NO_3^- concentrations in the drainage water from the lysimeters, which were irrigated with liquid fertiliser, were higher and were observed earlier than the corresponding ones for wastewater. This possibly indicates a temporary NH_4^+ adsorption to the soil particles, and higher denitrification due to the higher organic carbon of wastewater. The amount of C available to microorganisms is often the limiting factor for denitrification (Weier *et al.*, 1994), and this may indicate increased denitrification when wastewater is applied. On the other hand, significant differences between clay and sand were recorded for N-leaching and for N-uptake in shoots. Growth in clay lysimeters was much higher than that in sand lysimeters. Uptake of N in shoots in clay lysimeters was almost thrice that in sand. The N accumulated in the willow shoots in the clay lysimeters corresponded to 35% of the total amount of N supplied. The corresponding figure for sand was 12.5%.

Nitrogen processes following irrigation with wastewater

The amount of N leached is of course related not only to the biomass produced, but also to the other N processes that take place after fertilisation with wastewater. To increase denitrification is thought for instance to be one way of reducing the total amount of N-leaching (Snow *et al.*, 1999; Sakadevan, Maheshwari & Bavor, 2000). To obtain a better understanding of the N pathways, it is necessary to study the interactions between the various N processes. After irrigation with wastewater, N is partly accumulated in the above- and belowground plant biomass through growth, and can simultaneously be lost through leaching to the groundwater, through gaseous loss due to denitrification or by ammonia volatilisation. Nitrogen that is immobilised by microorganisms can later be mineralised and enter the soil pool. Additionally, N can enter the soil pool through litter decay. In the experiment of Paper I, only leaching and accumulation in the aboveground plant parts were measured. If the amounts of N entering the soil pool by litter decay, and the amounts of N incorporated in belowground plant biomass, are calculated by means of the conceptual model of Aronsson (2000), a theoretical assessment of N pathways in the lysimeters of Paper I can be obtained (Table 2).

Table 2. *Theoretical N budget (kg N ha^{-1} yr^{-1}) for each treatment in the lysimeters of Paper I, including the mean values (n=2) of N-application via irrigation-fertilisation and N leaching loads. The * values indicate values calculated on the basis of the conceptual model of Aronsson (2000). 'Not accounted for' values indicate the net result of N-mineralisation and N losses through volatilisation. WW = wastewater and LF = liquid fertiliser*

	Clay WW	Clay LF	Sand WW	Sand LF
Irrigation	+320	+320	+320	+320
Leaching	–80	–81	–90	–116
Shoots	–115	–110	–44	–38
Stumps*	–8	–5	–2	–2
Coarse roots*	–12	–9	–4	–3
Leaf litter to humus*	–96	–68	–30	–24
Fine-root litter to humus*	–115	–82	–35	–29
Not accounted for	+106	+35	–115	–108
Σ	0	0	0	0

In the budget of Table 2, the term 'N not accounted for' refers to the net result of mineralised N and to N lost to the atmosphere by denitrification or ammonia volatilisation. Denitrification is reported to be higher in clay soil, in water-saturated and warm soils, and when NO_3^- and C are available (Barton *et al.*, 1999; Smith & Bond, 1999; Hooda, Weston & Chen, 2003). In the clay lysimeters of Paper I, irrigation was conducted at high intensity (the saturation level was therefore high) during summer (soil temperature was therefore comparatively high). Furthermore, high levels of NO_3^- were present (owing to irrigation with wastewater and liquid fertiliser) and the organic matter content was higher than that in sand lysimeters. The organic content of the wastewater might have also contributed to increased denitrification. Denitrification in the clay lysimeters irrigated with wastewater could therefore have been substantial. Aronsson (2000) has estimated that denitrification of fertilised SRWC can reach at least 70 kg N ha^{-1} yr^{-1}. Denitrification in a coppiced *Eucalyptus globulus* Labill. plantation irrigated with wastewater reached 78 kg N ha^{-1} yr^{-1} in clay soil (Hooda, Weston & Chen, 2003). Smith, Freney & Bond (1996) found that ammonia volatilisation occurs in fields irrigated with wastewater, and that, although it is difficult to estimate, it is related to soil temperature, soil pH and evaporation. It is difficult to estimate the level of denitrification and ammonia volatilisation in the experiments of Paper I. It is also difficult to determine which process lies behind the 'N not accounted for', and to what extent. However, if the gaseous losses that might have occurred are considered—especially those from the clay lysimeters treated with wastewater—the N mineralised in the clay lysimeters could be much higher than the 106 kg ha^{-1} yr^{-1} calculated in the budget. This is a further indication that extensive N mineralisation may occur when fertilised SRWC is grown on a fertile site with large amounts of organic N (Slapokas, 1991; Aronsson, 2000). It may therefore be concluded that, when high irrigation rates with N-rich wastewater are applied to fertile agricultural soils, the high soil-moisture level and the organic content of the water may increase denitrification, while at the same time N mineralisation occurs, especially when the level of N already present in the soil is high. The uncertainty regarding the N pathways is considerable, and the risk of leaching is substantial.

Wastewater irrigation of SRWC at Enköping—Field situation

In the 75-ha SRWC plantation at Enköping, the soil is gyttja clay, *i.e.* similar to the clay soil in the lysimeters, and the concentration of N in the irrigation water is not as high as in the experiment described in Paper I. The N-rich wastewater, otherwise called supernatant, is stored in ponds in winter, during which a reduction of N has been observed (Aronsson, personal communication). Furthermore, the supernatant is usually mixed with conventionally treated wastewater during the irrigation period, as an additional water supply to the SRWC. The average concentration of total-N in the irrigation water for 2002–2004 was 64 mg l^{-1}, 43 mg N l^{-1} of which was in the form of NH_4-N (Klemedtsson *et al.*, 2005). The irrigation load in those years was on average 2.8 mm d^{-1}, with small variations from year to year. Irrigation took place from May to September. The total supply of N was on average 153 kg N ha^{-1} yr^{-1}, *i.e.* substantially higher than the recommended application rate for conventional fertiliser on SRWC, which is about 80 kg N ha^{-1} yr^{-1} (Ledin *et al.*, 1994).

Paper I showed that large amounts of N can be satisfactorily treated by irrigation of newly harvested willows, even at high irrigation rates and with wastewater containing a high N concentration, during a short period. The results from the irrigation at Enköping, which was carried out in a more realistic fashion, with moderate daily irrigation rates, showed that N-leaching was *ca.* 7 kg ha^{-1} yr^{-1} (Klemedtsson *et al.*, 2005). The concentrations of NO_3-N in the groundwater during the irrigation period were well below 10 mg l^{-1}, which is the limit for NO_3-N in drinking water. This agrees with the findings of Aronsson (2000), from

SRWC irrigated with wastewater in Sweden; Aronsson suggested that at least 200 kg N ha^{-1} yr^{-1} can be retained (plant uptake, immobilisation in the soil and gaseous losses) and 'treated' without contaminating the groundwater by N leaching. The growth of SRWC in Enköping was *ca.* 9 t DM ha^{-1} yr^{-1}, which is rather high compared to other commercial fields in the area (Nordh, personal communication). This indicates an absence of negative effects of wastewater irrigation on plant vigour. It is evident that the irrigation method in Paper I was unsuitable for practical field use; it was merely a means of quantifying N leaching under 'worst-case' conditions. Under real field conditions at Enköping, wastewater irrigation appeared to be applied in a sustainable manner. The *ca.* 150 kg N ha^{-1} yr^{-1} applied with the wastewater were treated satisfactorily, at least in terms of N-leaching to the groundwater.

What maximum sustainable N amounts and irrigation loads can be applied to SRWC?

Since N-leaching and reduced growth were not observed in the field at Enköping, it might be concluded that even higher N loads could be applied for a more cost-efficient N treatment (Rosenqvist *et al.*, 1997). This could be done by increasing either the irrigation volume or the N concentration in the wastewater. In the system at Enköping, which is based on a novel and rather sophisticated approach, it is possible to adjust the N concentration in the irrigation water by mixing it with treated wastewater. In simpler systems, however, which do not store or mix the wastewaters, N concentrations cannot be regulated by dilution, hence only the irrigation rate can be adjusted to affect the total amount of N applied.

Several models for wastewater irrigation of various short-rotation crops have been constructed (Ou *et al.*, 1997; Al Jamal, 2002). Most of them are based on the plant's water requirements and aim to optimise water use, but do not take into account the N content or the content of other hazardous compounds in the wastewater. A simple water-balance equation, used for deciding irrigation rates with wastewater, was given by Roygard *et al.* (1999), by the formula:

$$I = ET - P - \Delta S, \tag{1}$$

where I is wastewater irrigation (mm), ET is evapotranspiration (mm), P is precipitation (mm) and ΔS is the change in soil-water storage (mm). All of the components of the above equation vary greatly with geographical area, soil type, time period, plant species, *etc.* In particular, evapotranspiration varies from clone to clone of the same species (Persson & Lindroth, 1994). Therefore, for wider applicability, a more complex model would be required, because a range of conditions must be covered (Bond, 1998; Snow *et al.*, 1999). The present thesis does not aim to construct or to propose a model for estimation of wastewater irrigation levels on SRWC. It is, however, of interest to investigate the restrictions, but also the potential, of SRWC irrigated with wastewater, on the basis of the observations and assumptions of the previously mentioned examples, and by using simple approaches.

Table 3. *Overview of the experimental treatments and results (1999–2001) in Larsson et al. (2003). Wastewater = WW, Precipitation = Pptn*

Site	N in WW mg l^{-1}	WW applied 1×PE mm yr^{-1}	Pptn mm yr^{-1}	N loads kg N ha^{-1} yr^{-1}		Max N in groundwater mg l^{-1}		Growth yr 1-3 t DM ha^{-1} yr^{-1}	
				1×PE	3P×E	1×PE	3×PE	1×PE	3×PE
France	49	198	830	99	237	1.4	8.4	7	8
Greece	49	1124	282	414	1176	36	18	5	9.7
Sweden	4	455	507	18	53	0.3	0.5	6.6	7.3

Restrictions and potential of wastewater irrigation on SRWC—An example

Experiments on wastewater irrigation of SRWC were established in four European countries (France, Greece, Sweden, UK) within the European Union project FAIR5-CT97-3947 (Larsson *et al.*, 2003). There was a range of differences in soil and wastewater types, but the willow clone used was the same, *i.e. S. viminalis*, 'Jorr', which was developed for north-European climatic conditions. Similar methodology and calculations as in equation (1) were used to decide irrigation levels for the different treatments. 'Potential Evapotranspiration' (PE) was calculated by multiplying a crop coefficient which varies throughout the growing season (and which took local climatic conditions into consideration), by the Penman evaporation rate (Lindroth & Båth, 1999). Irrigation at the rates 2×PE and 3×PE, and a control with tapwater, were included. Details of the experimental treatments and results are given in Table 3 (above).

A comparison between the sites in France and Greece is of interest, since the water had approximately the same N concentration (*ca.* 50 mg N l^{-1}), even though the origin of the wastewater differed. In France, it was derived from a chicory-processing industry; in Greece it was primary effluent from a municipal wastewater treatment plant. The PE varied greatly between these locations, hence the N loads applied also varied widely. This difference was reflected in the NO_3-N concentration in the drainage water; in France it was well below 10 mg l^{-1} for all treatments and years, *i.e.* much lower than the limit for drinking water in the EU. In Greece, NO_3-N concentrations in the groundwater were higher, and the highest observation was for 1×PE in early spring. Later in the summer, the NO_3-N concentrations were much lower. In Sweden, the N content of the wastewater was very low and the N amounts, which were decided on the basis of PE, were lower than the SRWC requirements in terms of N. No leaching was observed in Sweden, and biomass production for 3×PE was the lowest among all locations (Table 3). The N loads applied in Greece (1176 kg ha^{-1} yr^{-1} for 3×PE) were greater than the plants required, and probably much larger than the amounts that can be retained by SRWC. The plants did not suffer from water deficits—clearly the main hazard for a SRWC plantation in south-European climates—and N-leaching was observed. Even though SRWC grown in warmer climates might be able to treat and utilise larger amounts of N owing to higher denitrification and to higher potential growth, the total N-load applied with wastewater must be also taken into account, for sustainable application to SRWC. Differences in growth were also observed, and were related either to N loads or to other local conditions; in Sweden the low level of N applied led to low biomass production, and in Greece SRWC growth varied greatly between treatments, and was greater for the higher rate of irrigation with wastewater, although the clone was not suitable for the local conditions. In France, the lower than expected biomass production was attributed to poor soil conditions, and to pest attacks which damaged the plants (Larsson *et al.*, 2003).

Factors to consider for sustainable irrigation of SRWC with municipal wastewater

The experiments described above indicate the potential of wastewater irrigation on SRWC in different geographical areas and with different types of wastewater. They also indicate several factors which must be carefully considered if sustainability is to be achieved in short-rotation plantations irrigated with wastewater. Species or clone selection is of great importance. The plant material must be adapted to the specific climatic conditions, in order to produce more biomass, and must also tolerate pest attacks. Furthermore, when it is treated with high irrigation loads, the plant species or variety with the highest evapotranspiration rate should be used: the greater is the evapotranspiration, the more water is 'filtered' by trees and the smaller is the potential hazard due to leaching. The climatic conditions of a specific area must be also taken into account. The precipitation load is an important factor to be considered when deciding the wastewater irrigation levels, and for avoiding excessive leaching of N or of other compounds, since N-leaching and drainage to the groundwater are often correlated (Bergström, 1995). The soil type in a SRWC irrigated with municipal wastewater is of great importance. Soil N processes vary with soil type, and are critically important in deciding the N loads applied with the irrigation. Denitrification rates in soils irrigated with wastewater, although they are difficult to estimate, appear to be higher in clay than in sandy soils. Since NO_3^- is not limited, anaerobic microsites—a prerequisite for denitrification—are more likely to occur in fine-textured than in coarser soils (Barton *et al.,* 1996). Furthermore, fine-textured soils (clayey and silty) have better water-holding capacity than coarser soils; larger wastewater amounts can therefore be treated. However, this thesis cannot answer with certainty the question posed above, 'What are the maximum sustainable N amounts and irrigation loads applied to SRWC?' Nevertheless, if all N-processes could be quantified, *i.e.* N-uptake in shoots, net N-mineralisation, denitrification, NH_3 volatilisation and N-leaching, which depend greatly on site-specific characteristics, then a better understanding of a sustainable application of wastewater to SRWC would be obtained.

As mentioned above, the sustainability of a SRWC phytoremediation system irrigated with wastewater is judged not only by the N amounts leached or by other potential environmental hazards, but also by the growth achieved. Therefore, the planning of irrigation must aim to attain not only the least damage, but also the best growth of SRWC. When, instead of municipal wastewater, other wastewater types are used to irrigate a SRWC, the irrigation load should be adjusted according to any hazardous compound present in the water, to avoid growth reductions. One example of such potentially problematic wastewater is landfill leachate.

Irrigation of SRWC with landfill leachate

The water that originates from the percolation of water through, and the generation of water from, material in landfills, is called landfill leachate. The composition of the leachate is highly variable, and is influenced by the type of solid waste deposited, the age of the landfill, the rate of waste input, and the climate (Christensen *et al.*, 2001; Van der Sloot *et al.*, 2003). In addition to a high concentration of NH_4-N, and taking into account that N is a plant nutrient, there is a vast range of hazardous compounds in the leachate. A list of dangerous compounds in landfill leachates, and average values for Swedish landflill leachates, is given in Table 4 (Alker, 1999; Öman, 2000). The quality of landfill leachate, in terms of nutrient requirements for SRWC, is inferior by comparison with municipal wastewater, since P is limiting (Table 1). Furthermore, the high ionic strength, due to high salt concentrations, and the high content of organic compounds in landfill leachate, may negatively affect SRWC growth after irrigation, notwithstanding the beneficial presence of NH_4-N. Therefore, and especially when the irrigation level of landfill leachate on SRWC is based on potential evapotranspiration, toxic effects on the plants may occur, and SRWC growth will decrease (Cureton, Groenevelt & McBride, 1991; Alker, 1999; Stephens, Tyrrel & Tiberghien, 2000). Consequently, transpiration will be lower and the treatment of compounds in landfill leachate may be ineffective. In addition, the production of biomass, which is an important end-product, will be reduced.

Table 4. *Typical chemical composition of landfill leachates (Alker, 1999) and Swedish averages for the respective compounds (Öman, 2000). COD is the Chemical Oxygen Demand and BOD_5 is the Biological Oxygen Demand after five days*

Parameter	Typical leachate range	Swedish average values
pH	5.3 – 8.5	7.6
COD (mg l^{-1})	150 – 10000	750
BOD_5 (mg l^{-1})	100 – 90000	26
TOC (mg l^{-1})	10 – 25000	260
NH_4-N (mg l^{-1})	1 – 1500	340
Tot. N (mg l^{-1})	1 – 2000	350
Tot. P (mg l^{-1})	0.1 – 30	1.3
Cl (mg l^{-1})	30 – 4000	1600
Na (mg l^{-1})	50 – 4000	510

The identification of the irrigation load which will not harm or decrease the growth of SRWC in a field situation is therefore essential. This load is related to the chemical composition of the leachate, the climate, the soil conditions, the irrigation method and the plant material selected, and thus is site-specific. The leachate in Paper II was taken from a landfill in Högbytorp, central Sweden. At that site, the leachate is collected and aerated in lined ponds for reduction of N, *i.e.* nitrification–denitrification, and is then pumped to a 6.5 ha SRWC for irrigation. Growth was poor in that plantation after the first year of irrigation (Fig. 1, below), and the plants exhibited stress symptoms (Dimitriou, Aronsson & Weih, 2003).

The experiment of Paper II aimed to identify problems related to the poor performance of SRWC in the field at Högbytorp.

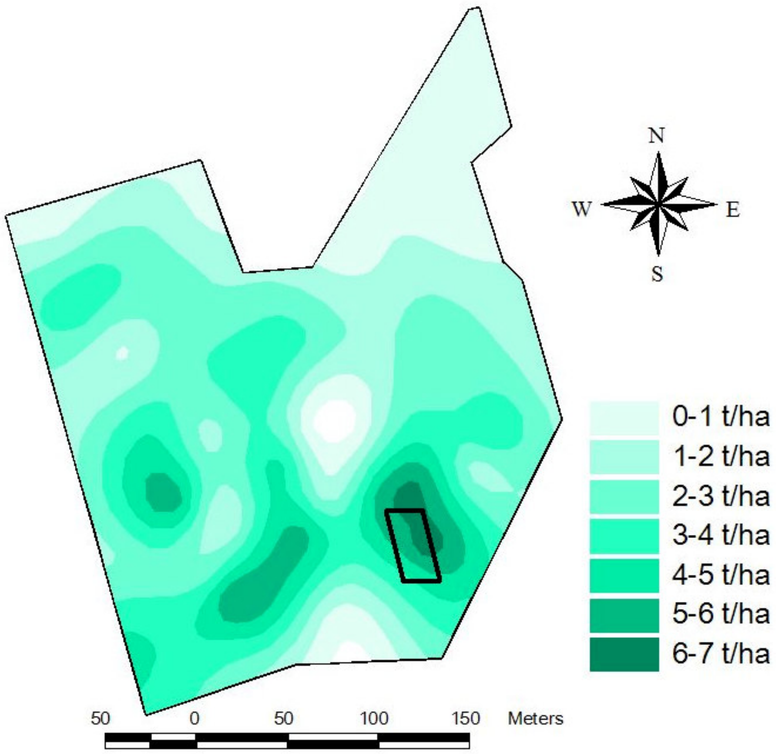


Figure 1. Distribution of the shoot DM in the willow plantation at Högbytorp, based on smoothed data from 52 sampled row-sections. The rhomboidal area corresponds with the non-irrigated reference area.

Results from the pot-trial (Paper II)

The concentration of NH_4^+ in the leachate used in Paper II was lower than that in Paper I. In spite of the differences in experimental conditions (pots–lysimeters, cuttings–harvested plants), plants in pots irrigated with leachate did not appear to suffer from toxic effects due to NH_4^+. The concentrations of NH_4^+ for the 'stronger' leachate treatment were *ca.* 90 mg l^{-1}, lower than those in similar experiments in the literature (Alker, 1999; Watzinger *et al*., 1999), and damaged plant parts due to NH_4^+ toxicity were not observed. Furthermore, although N concentrations in stems for the 'strongest' treatment were higher than those in the control (14.4 ± 1.5 and 6.4 ± 0.1 mg g^{-1} DM, respectively), the concentrations were not exceptionally high compared with those given in the literature (Aronsson, 2000). The relative growth rate (RGR) of the control plants was also significantly higher than that of the leachate-irrigated plants (0.537 ± 0.016 and 0.360 ± 0.017 wk^{-1}); this may also have contributed to the higher N concentrations in the leachate-treated plants, owing to the dilution effect. Other compounds in the leachate may therefore have been responsible for the decrease in growth.

The concentration of Cl^- in the leachate used in the experiment of Paper II was 723 mg l^{-1} and was considered too high to be used undiluted in an experiment with young plants grown in pots. According to FAO guidelines for the quality of irrigation water in agriculture, there are severe restrictions on the use in agriculture of waters that contain >350 mg l^{-1} Cl^- (FAO, 1985). For landfill leachate, a suggested application range reported by Alker (1999) lies between 56.8–152 mg Cl l^{-1}. The leachate used in Paper II was therefore diluted to 240, 180 and 120 mg l^{-1} for the different treatments. Symptoms of phytotoxicity have been reported in a number of related studies, and were related in most cases to the high ionic strength due to high NaCl concentrations (Ettala, 1988; Cureton, Groenevelt & McBride, 1991; Chan, Wong & Whitton, 1999; Stephens, Tyrrel & Tiberghien, 2000). It must be noted, however, that in several of the abovementioned studies the leachate was undiluted and 'stronger' than that used in our experiments, and contained higher salt concentrations; its toxicity even resulted in dead plants. It is generally unclear whether the plants are affected only by the actual concentration of salts or by other factors, such as the cumulative effects of a number of toxic elements. The concentration of Na in the leaves of the leachate-irrigated plants in Paper II was 1112 μg g^{-1}, and was significantly higher than that in the control (658 μg g^{-1}). Although obvious toxic effects did not appear in our study, growth reductions in the leachate-irrigated plants indicated that a negative effect was present. The experiment can neither answer the question which component caused the growth reduction, nor can it reveal which concentrations of Cl^- in the water were harmless to the plants. There were, however, indications that in addition to the high ionic strength of the leachate, its low P concentration (4 mg l^{-1}) may also have affected growth, since P concentrations in leachate-irrigated plants were much lower than those of the control. However, had more P been applied with higher irrigation of landfill leachate, the P stress might have been lower; but at the same time, the applied Cl^- amounts would also have been higher. Thus it is difficult to assess how a stronger leachate would affect the plants.

To quantify stress, relative growth data were used, but growth is impractical as a rapid indicator of stress. In contrast, leaf length and leaf fluctuating asymmetry are morphological characteristics that can be easily and rapidly measured under field conditions. Leaf fluctuating asymmetry did not differ between different treatments. On the other hand, leaf length was significantly correlated with relative growth rate in Paper II. Furthermore, in the leachate-irrigated plants, leaf length differed significantly from that in the control, and showed promise as a diagnostic tool for stress. This was therefore also tested in the field.

Field observations

To monitor some of the effects of leachate irrigation on SRWC at Högbytorp, growth, leaf length and other plant traits were also measured in the field during the growing season. The 6.5-ha SRWC plantation adjacent to the landfill was irrigated with leachate by means of sprinklers (Dimitriou, Aronsson & Weih, 2003). The plants had grown for one year and were harvested during the previous winter (one-year-old shoots on two-year-old roots). A non-irrigated 0.1 ha plot was used as a reference. The average daily leachate load was 2.6 mm, which resulted in an accumulated load of *ca.* 260 mm, equivalent to about 530 kg N-tot ha^{-1}. The chemical composition of the leachate was similar to that of the undiluted leachate used in the pot experiment (Paper II). Figure 1 illustrates biomass production (t DM ha^{-1}) on the field during the first year of irrigation.

Production on the reference area was 5.7 t DM ha^{-1}, which is considered high for one-year-old shoots, and was significantly higher than that on the area irrigated with leachate (2.4 t DM ha^{-1}) (Dimitriou, Aronsson & Weih, 2003). Since there was only one reference area, it is possible that the differences in growth were due to factors other than irrigation with landfill leachate. At the first sampling, one week after the start of irrigation, there were no differences between the irrigated and non-irrigated parts of the field, as measured in terms of leaf length. At the second sampling, three weeks later, significant differences in leaf length between the irrigated and the reference area were observed ($p<0.021$). This indicates that irrigation could have been responsible for the growth differences during the season, and reinforces the conclusion that leaf length is an appropriate, rapid indicator of stress, which can be used under field conditions.

The Na concentration in shoots of SRWC collected from the leachate-irrigated area at Högbytorp, after the end of the growing season, was 419 mg kg^{-1}. Although no data are available from the reference area, data from several plantations elsewhere in central Sweden (Vigre & Ledin, 1993, unpublished data) suggest that Na concentrations on the leachate-irrigated area were about ten times higher. The respective P and N concentrations in the plant parts were normal. It may therefore be inferred that the high amounts of salt applied, in the form of NaCl, strongly affected growth, owing to the high Na concentration in shoots. Sprinkler irrigation may also have contributed to the poor growth, since direct contact between leaves and leachate caused leaf necrosis in some parts of the field (observations at Högbytorp).

The sustainability of landfill leachate irrigation on SRWC

It is uncertain what N load may be applied to a SRWC by irrigating it with any type of wastewater, without contaminating the groundwater (see comments above, concerning municipal wastewater). However, the 530 kg N ha^{-1} yr^{-1} applied to the field at Högbytorp was high, and N-leaching was substantial (based on N concentrations in drainage water from the field). This can be attributed to the large amounts of N applied, but also to the poor growth of the SRWC due to irrigation with leachate, which resulted in less plant uptake of N and less C input to the soil from fine-root litter. This in turn probably resulted in lower denitrification rates (Luo, Tillman & Ball, 1999; Hooda, Weston & Chen, 2003). Because of the rather high level of organic compounds in the leachate irrigation (TOC *ca.* 330 mg l^{-1}), the risk that organic carbon compounds may be leached to neighbouring water systems should also not be neglected. To avoid having to consider leaching to the groundwater as a threat to the sustainability of the system, SRWC can be grown on covered areas of landfills which are no longer in use. In most landfills in Sweden, such parts are lined and are impermeable at the base, and can therefore reduce leaching to deeper layers (Ettala, 1988; Nixon *et al.*, 2001; Hasselgren, 2003).

All parts of the irrigated SRWC field at Högbytorp received the same amounts of leachate. The wide variations in growth within the leachate-irrigated area, illustrated in Figure 1, indicate that the composition of the leachate is not the only factor responsible for differences in growth performance, and that other factors may also be responsible. Soil type and site topography probably play a part in growth performance, in addition to the amounts of leachate used. Nevertheless, the dilution of leachate or a decrease in the total amount of irrigation could have contributed to better growth of the SRWC. Fertilisation with P and micronutrients would possibly have resulted in better growth, but it was most likely the high-saline character of the water that contributed to the poor growth. Therefore, dilution of the leachate prior to irrigation, and additional fertilisation, might promote SRWC growth and contribute to the avoidance of negative effects on SRWC growth. Furthermore, the selection of a salt-tolerant willow clone, which can also produce large amounts of biomass, would result in better treatment of leachate and better SRWC performance. In Paper II, it appears that there are considerable genetic differences as regards these traits, since the clones 'Jorr' and 'Loden' performed better in terms of growth than the other clones tested. Furthermore, to improve sustainability by avoiding growth reductions, the rapid identification of stress in a field would help in the adjustment of the irrigation rate of SRWC. Leaf length of SRWC could be used as such a tool.

Composition of log-yard runoff

Compared with municipal wastewater and landfill leachate, the concentration of various compounds in log-yard runoff is quite low (Table 2 in Paper IV and Table 1). Phenolic acids, which are potentially toxic if they occur at high concentrations in water, usually have a higher concentration in runoff from log-yards where hardwood is stored, compared with Norway spruce (*Picea abies*) as in Papers IV and V (McDougal, 2002). Oxygen-consuming organic matter is considered to be the most serious potential threat when runoff is directly discharged to nearby water systems (Zenaitis, Sandhu & Duff, 2001). TOC concentrations vary greatly, depending on the type of runoff and the storage regimes; in Papers IV and V, TOC was 191 mg l^{-1} and 56 mg l^{-1} for recycled runoff and runoff from an open system, respectively. The TOC concentration in Swedish landfill leachate is typically *ca.* 200–300 mg l^{-1} (Öman, 2000). This is a further indication that TOC in log-yard runoff should not pose a threat if it is used to irrigate SRWC.

What problems arise when log-yard runoff is used to irrigate SRWC?

The sprinkling of sawtimber and pulpwood with water during summer affords protection from damage by insects and fungi, and from cracks due to the drying-out of the wood. Even without sprinkling, wastewater is still produced in large quantities after rainfall or snow-melt. At Heby sawmill, a medium-sized sawmill in central Sweden at which the experiment in Paper V was conducted, *ca.* 100,000 m^3 water are consumed annually for watering the stored wood. This results in large quantities of runoff during the summer; *i.e. ca.* 60,000 m^3 yr^{-1}. Runoff was formerly discharged directly to the nearby river, before it was decided by the log-yard operator that it would be collected and used to irrigate two adjacent fields with naturally grown couch grass and SRWC, respectively. In this way, it was intended that the amount of runoff would be reduced by evapotranspiration and filtered, before being naturally drained to the groundwater and onward to the river.

It was decided that both the SRWC and couch grass field would be irrigated by sprinklers during the growing season. The field with couch grass is on a slope and has an area of *ca.* 0.5 ha; the irrigation rate was *ca.* 66 mm d^{-1}. The area of SRWC used for irrigation was only 1 ha, which gave rise to irrigation rates of *ca.* 4000–4500 mm yr^{-1} in the year 2002 (33–38 mm d^{-1}). These are very high rates compared with those for municipal wastewater and landfill leachate, and led to parts of the field being flooded during the entire summer. In spite of this, the plants survived, but their growth was poor (observations only; no measurements were made).

Experiments with log-yard runoff (Paper IV and V)

The experiments in Papers IV and V were carried out in an attempt to mimic irrigation intensity in the ‘real’ situation conducted at Heby, and to estimate whether the irrigation method chosen was sustainable. The ability of various species, grown in different types of soil, to achieve sustainable remediation of two types of runoff was also investigated. The species selected were *Salix* spp (commercially grown as SRWC), *Alnus glutinosa* (N-fixing) and *Elymus repens* (couch grass, naturally occurring at Heby). An additional experiment was set up at Heby in the irrigated couch grass field, to evaluate its retention capacity and the quality of the groundwater, after irrigation with runoff produced in the nearby log-yard.

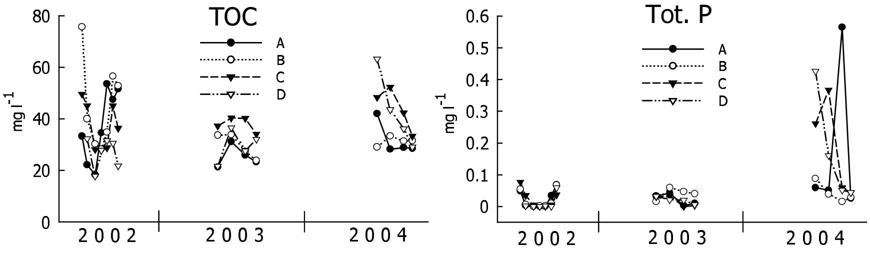
A major difference between the experiments in Papers IV and V and the lysimeter experiment in Paper I was the irrigation intensity. The daily irrigation loads imposed on the lysimeters in Papers IV and V were 10, 20 and 49 mm, respectively. Retention in the lysimeter experiments was high for all irrigation levels, soil types and plant species used. The relative retention found in Paper IV was >90% for TOC, >50% for phenols and >50% for P, and in Paper V was >45%, >75% and >75%, respectively. The phytoremedial efficiency of SRWC depended neither on plant species nor on soil type: the key factor was irrigation intensity (Paper IV). The relative retention of TOC and P in all lysimeters was higher at the lower irrigation intensity. Phenols, by contrast, were retained most strongly at the highest irrigation rate. However, the actual phenol concentrations in the groundwater were low (*ca.* 0.1 mg l^{-1} on average, for both low and high irrigation rates in Paper IV, and *ca.* 0.1 mg l^{-1} in Paper V), and future hazards were not expected.

Since the retention capacity was correlated with irrigation level in the lysimeter experiments, it is of interest to discover whether this also applies under field conditions. Although the concentration of P in log-yard runoff was only 1.43 mg l^{-1}, daily irrigation with 66 mm resulted in a total annual volume of 7,200 mm, consequently in a very high total P load applied to the Heby field. The total amounts of TOC and P applied were *ca.* 5,000 and 100 kg ha^{-1} yr^{-1}, respectively. During the first year after irrigation, TOC and P in the groundwater were not alarmingly high (Fig. 4, Paper V), but a sustainable irrigation regime would require these amounts to remain fairly low (SNV, 2003).

Suggestions for sustainable application of log-yard runoff

In Figure 2, the concentrations of TOC and P in the groundwater at the couch grass field at Heby for a three-year period are illustrated. The TOC concentrations for the second year of irrigation with runoff were lower, and for the third year were at approximately the same level, than those in the first year, and no major variation during these years were observed (Fig. 2). For P, concentrations in the second year did not differ substantially from those in the first year, and were generally low. An increase in tot. P was observed, however, during the third year of irrigation, but at the end of the season the tot. P values were close to zero (Fig. 2). If the lower irrigation levels applied to SRWC (*ca.* 33–38 mm) are considered, then a more sustainable treatment of log-yard runoff would probably be achieved. Nevertheless, a potential harvest of SRWC, which produces *ca.* 6–10 t DM ha^{-1} yr^{-1}, results in a potential output of *ca.* 6–10 kg ha^{-1} P yr^{-1}, compared to the 100 kg P ha^{-1} yr^{-1} applied. Therefore, a decrease in the irrigation level and an increase in the area of SRWC, to reduce potential future hazards, would be considered a step towards sustainability, when it is considered that TOC and P, when in excess, are respectively responsible for low oxygen levels and the eutrophication of surface waters.

Figure 2. Mean concentrations (n=2) of TOC and total P in groundwater sampled within (A-B) and downhill (C-D) the couch grass field (Paper V) that was intensively irrigated with log-yard runoff during three seasons (2002-2004). The figure is partly based on results presented by Jonsson (2004).



SRWC fertilised with sewage sludge and wood-ash

In the experiments in Paper III, the residues used on SRWC were not in liquid form, as in the previously described experiments, but in solid form. In Sweden, fertilisation of SRWC with sewage sludge is a rather common practice, and about 10,000 ha (*ca.* 60% of the total SRWC plantations) are fertilised with sludge after harvest (Slagbrand, personal communication). Sludge contains some N (mainly organically bound) and large amounts of P, but very little potassium (K); wood-ash contains some P and large amounts of K (Table 1). Wood-ash, when available, is mixed with sludge, since it contains some P but also K, and a more balanced fertiliser is produced. The mixture is applied to SRWC as a replacement for conventional inorganic fertilisation. According to legislation, the amount of P in sludge–ash applied every 3–7 years should not exceed 22–35 kg P ha^{-1} yr^{-1}, depending on the original P-status of the soil (SNV, 1994).

Sludge and ash contain, besides nutrients, a range of heavy metals that can accumulate in soil. SRWC is a non-food, non-fodder crop, and therefore no direct entry of metals to the food chain is to be feared. However, the future cultivation of food-crops on fields fertilised with a sludge–ash mixture, might lead to elevated levels of heavy metals in the agricultural products. The reported ability of willow to take up heavy metals (particularly cadmium, Cd) and nutrients in excess (Landberg & Greger, 1996; Perttu & Kowalik, 1997), might reduce potential risks to sustainability. The idea is that the heavy metals applied with sludge–ash mixtures on SRWC would be decreased by plant uptake and retention in the soil-plant system. At harvest, stem parts which contain heavy metals would be removed, burnt and partly recycled in SRWC with any future ash application. The ash that is returned to SRWC is bottom-ash, since it has a lower heavy-metal content than fly-ash. In this way, a sustainable recycling system, involving energy biomass production, the treatment and utilisation of nutrient-rich residues, and the minimisation of heavy-metal accumulation in the soil, would be achieved.

Results from field experiments (Paper III)

The experiments in Paper III were carried out in the county of Uppland, central Sweden, to investigate the fate of heavy metals after the application of legally permitted sludge–ash mixtures for fertilisation of SRWC. For comparison, a treatment with twice the maximum permitted sludge–ash amount was also tested. Sludge and ash were taken from the local wastewater treatment plant and power plant, respectively. Paper III showed that the application of different sludge–ash mixtures resulted in changes of soil pH in the topsoil that had no effect on heavy-metal uptake in plant parts. It was proposed that Cd, the most soluble of the metals in the soil, and considered the most hazardous to human health, would decrease in SRWC stems in the ash treatments. A decrease in the phytoremedial effect of SRWC was therefore to be expected. This proved not to be the case, and a potential output of Cd in the biomass removed would occur after three years' cultivation. For the other metals, a potential output *via* harvest would not compensate for the input in sludge–ash mixtures. Consequently, the amount of these metals would increase in the soil in all treatments. In addition, shorter harvest intervals appeared to favour the remedial ability of SRWC. Nevertheless, the difference between the amount of metals supplied to the soil in the sludge–ash mixture, and the potential output in harvested biomass after SRWC harvesting, were within the permitted limits for sludge applications. It can therefore be concluded that application of the legally permitted sludge–ash mixtures can be suggested as an alternative method for fertilising SRWC, and that SRWC reduces Cd concentrations in soil.

How would SRWC increase the removal of metals?

The remedial effect of SRWC should combine high metal concentrations in stems with high rates of biomass production. Therefore, if the level of both of those factors was increased, metal removal would also be greater. Larsson (1998) presented comparisons between 'old' willow clones and new commercial clones more recently introduced on the market. The production differences between the clone used in our experiments (*S. viminalis*, '78021'), and clones currently in widespread use in Sweden, were up to 53%. The use of new clones could therefore lead to an increased phytoremedial effect of willow. In many cases, and especially when sludge only is applied, additional fertilisation with inorganic fertilisers might contribute to better growth (Labrecque & Teodorescu, 2003). In Paper III, however, the sludge–ash treatments showed no differences compared with the control, which was fertilised with NPK fertiliser; therefore it is possible that growth would not be increased by conventional fertilisation.

In the experiments of Paper III, the concentrations of all metals in SRWC stems, including Cd, were low in all treatments as compared to values reported in the literature (Riddell-Black, 1994; Landberg & Greger, 1996; Klang-Westin & Perttu, 2002; Pulford, Riddell-Black & Stewart, 2002; Hammer, Kayser & Keller, 2003). This was probably related to the comparatively small amounts of heavy metals supplied to SRWC in the sludge–ash mixtures, and possibly also to site-specific conditions, *i.e.* the high clay content of the soil. The experiments of Paper III showed that concentrations of all metals in bark are far higher than those in the wood of the stems. The bark:wood ratio decreases with age and therefore shorter harvest intervals might contribute to higher amounts of metals removed, as was indicated in Paper III. Differences in the bark:wood ratio between willow clones of the same age have also been reported (Oliveira Miguel, 2003). Furthermore, clonal differences, in terms of preference for different heavy metals, have been reported by Pulford, Riddell-Black & Stewart (2002). In cases such as that of Enköping, where a particular metal, *e.g.* Cu, is the main threat to sustainability, clones which show 'preferential' uptake for that element should be selected for cultivation.

In the fields dealt with in Paper III, however, the high Cu concentrations in the sludge would still be responsible for high Cu amounts supplied in soil. The average concentration of Cu at Enköping for the year 2000 (the year in which the sludge in Paper III was used), although it varied throughout the year, was 702 mg kg^{-1} DM (Walgeborg, personal communication). The average Cu concentration in sludge from 48 municipal wastewater treatment plants in Sweden was reported as 390 mg kg^{-1} DM (Eriksson, 2001). Therefore, in many cases, as at Enköping, improved sludge quality would contribute to better sustainability after application on SRWC.

Sustainability and heavy metals in soils supplied with sludge–ash mixtures

The quality of soils which will be used for agriculture by future generations, must be considered when investigating whether fertilisation of SRWC with sludge–ash mixtures is also sustainable in terms of heavy metals over time. Therefore, it is of interest to compare the amounts of heavy metals in sludge–ash mixtures supplied to a SRWC, with the total amounts present in the topsoil. For SRWC plantations, where sludge is applied on the surface, and where in most cases no tillage is practised, a comparison with the total amounts in the upper 0–15 cm of the soil is relevant. The calculations in Table 5 refer to the field studied in Paper III (Lundby), and to the same sludge and ash as in Paper III. The increase (%) in the total amount of heavy metals after 'sl+ash' application over a 25-year period, *i.e.* over the economic lifespan of a SRWC plantation, is presented in the third column of Table 5 (see also Table 7, Paper III). The fourth column is the increase in the amount of heavy metals in the upper 0–15 cm of the soil (%), if SRWC were grown on the field and harvested every third year (*cf.* Paper III). Finally, in the last two columns, the number of years needed to double the total heavy-metal amounts in the upper 0–15 cm after 'sl+ash' is calculated, in the absence and presence of SRWC, respectively. For metals with a stem concentration (Paper III) below the detection limit, comparable values are given.

Table 5. *Total metal amounts in the soil at Lundby, and the calculated increase (%) in the 0–15 cm soil layer after 25 years for 'sl+ash', without or with SRWC cultivation. The last two columns show the number of years required to double the total amounts of metal in the soil, if 'sl+ash' were applied at Lundby, without or with SRWC cultivation, respectively*

	Total in 0–15 cm (kg ha^{-1})	Change in total metal amounts in soil after 25 years' application of 'sl+ash' (%)		Number of years to double the total amounts in soil after 'sl+ash'	
		no SRWC	with SRWC	no SRWC	with SRWC
Cd	0.36	+8.3	–26	+300	***–95***
Cr	92.6	+0.81	<+0.81	+3068	>+3068
Cu	59.1	+12.8	+12.2	+195	+204
Ni	53.4	+0.85	+0.51	+2966	+4854
Pb	43.3	+0.92	<+0.92	+2706	>+2706
Zn	225	+5	+3	+501	+833

On the basis of the theoretical calculations in Table 5, it can be concluded that Cu gives rise to most concern for sustainability in connection with sludge–ash application at Enköping. After 25 years with the legally permitted 'sl+ash' applications, the Cu concentration would increase by *ca.* 12%, *i.e.* the time required to double the soil Cu content would be *ca.* 200 years. However, if the Cu concentrations in residues from the Enköping wastewater treatment plant were lower, and were equal to the Swedish average (390 mg kg^{-1} DM), given the same

SRWC production and the same stem concentration of Cu, the increase in the 0–15 cm soil layer after 25 years would be *ca.* 7% (calculations not shown). In that case, 353 years would be required to double the amount of Cu in the 0–15 cm soil layer in a SRWC field (*cf.* Table 5—204 years). For the other heavy metals, increases after 25 years are very low compared to the amounts of metals already present in the soil. For Cd, a decrease in the total amount was calculated. After 25 years, and considering the increases in metal amounts in the soil after application of sludge–ash mixtures to SRWC, the sustainability of the system does not seem to be under threat for all metals, with the possible exception of Cu. Since the definition of sustainability refers in general to threats to future generations, it is by no means clear how many years it would take for possible future negative effects to affect sustainability. Given a SRWC generation length of 25 years, it would require *ca.* 8 human generations to double the amount of Cu in the topsoil, and *ca.* 14 human generations if the sludge were of average quality—and this only if a sludge–ash mixture were continuously applied during the entire period.

From Table 5, it is evident that SRWC at Lundby did not contribute substantially to the reduction of the total metal amounts in the soil. For all metals except Cd, the future amounts in the soil would not be notably decreased by removal in harvested biomass. Only Cd would be reduced in the soil after a SRWC harvest. In fact, if SRWC were grown at Lundby, it would theoretically take *ca.* 95 years to remove all Cd from the soil there, even if sludge–ash were applied. This is very important, since the exchangeable fraction of Cd in soil is between 10-40% (Eriksson, 1989), in contrast to *e.g.* Cr and Pb, which are highly immobile and potentially not as hazardous to future plantations (Eriksson, 2001). On the other hand, Cu, Ni and Zn are in a form such that they are likely to be less mobile under the specific conditions at Lundby, with a high clay content and high pH (McBride, Richards & Steenhuis, 2004). Furthermore, Cu and Zn are plant micronutrients, and an increase in the soil does not necessarily mean a high potential hazard level. Therefore, the fact that Cd—which is considered the most hazardous heavy metal to human health—is taken up by SRWC and that total Cd amounts in soil are reduced, is of great importance for increasing sustainability.

Is P a problem for sustainability?

Although the experiments on which this thesis is based did not explicitly address the problem of P, it cannot be ignored in any disscussion of sustainability. As mentioned above, sludge applications are partly regulated on the basis of the P supply; 35 kg ha^{-1} yr^{-1} for ammonium lactate-acetate soluble P (P-AL) I and II (<4 mg P-AL per 100 g soil DM), or 22 kg ha^{-1} yr^{-1} for P-AL III or higher (>4.1 mg P-AL per 100 g soil DM; SNV, 1994). According to Eriksson (2001), about 85% of Swedish agricultural land, and almost 100% of land on which sludge fertilisation is most likely to occur, is of class P-AL III or above. For practical reasons, sludge applications occur only during the establishment phase or after harvest of SRWC. Thus 88 kg P ha^{-1} are applied in a single dose to a SRWC plantation which will be harvested four years later. According to Ledin *et al.* (1994), the recommendations for P supply with inorganic fertilisers on SRWC are not clear, and lie between 20–

88 kg ha^{-1} for one cutting cycle. The authors suggest, however, that the applied P amounts should be closer to the lower values mentioned, for economic reasons, since P is easily adsorbed and becomes unavailable. For other arable crops, the recommendations for P are *ca.* 10 kg ha^{-1} yr^{-1} (Jordbruksverket, 2004), and still are much lower than the amount supplied with sludge–ash on SRWC.

A SRWC plantation fertilised with sludge–ash for 25 years would receive more than 500 kg ha^{-1} P. If the biomass production were *ca.* 10 t ha^{-1} yr^{-1} and P concentration in stems were 1 mg kg^{-1} DM, then *ca.* 100 kg P would be removed from such a SRWC plantation at stem harvest. The difference is still large, and the excess P might be lost to surface waters or leached to the groundwater. Nevertheless, losses of P *via* leaching or runoff have not been correlated with the P amounts applied with sludge, and are reported to be lower compared with other agricultural P amendments at similar P supply rates (Withers, Clay & Breeze, 2001; Elliott, O'Connor & Brinton, 2002). The origin of the sludge, which affects the bioavailable P content (Frossard *et al.*, 1996; Shepherd & Withers, 2001), and the soil type, which are linked to the transport mechanisms of P through the soil, and the subsoil properties responsible for P sorption (Djodjic, Borling & Bergstrom, 2004), are reported to be of greater importance to P losses than the total amounts applied.

Opinions regarding the risk of P losses after sludge application are rather contradictory (Dentel, 2004; O'Connor *et al.*, 2005), this inconsistency is reflected in the limits set by the legislation in different countries. The permitted supply of P by sludge fertilisation is far higher in other European countries than in Sweden, and in the USA, sludge applications basically depend rather on N amounts than on P, although concerns about the P supply have been raised (Morsing, 1994; Shober & Sims, 2003). A supply of P based on plant requirements would imply a more sustainable approach. In that case, however, the plant-available P in the soil would be more appropriately used as a regulator of the P supply when applying sludge. Plant-available P in sludge is approximately estimated to be *ca.* 50% of the total P supplied, and is strongly related to the origin of the sludge (Shober & Sims, 2003). Furthermore, Frossard *et al.* (1996) showed that the amount of sludge P taken up by ryegrass (*Lolium perenne* L.) was significantly and positively correlated both to the available P supplied *via* the sludge and to the soil's content of available P; however, these relationships applied only for a clay soil, whereas no such relationship was observed in a loamy soil, because of its higher content of available P. The soil's content of available P, and soil type, must therefore also be taken into account if plant-available P is to be used to regulate sludge applications. The concept would be even more complex if a sludge–ash mixture were applied, as was done in Paper III; there, uncertainty about the amount of plant-available P would be greater still. More research into the P supply after the application of sludge–ash mixtures on SRWC is therefore needed, for it to be possible to assess the sustainability of such a practice.

The 22 kg P ha^{-1} yr^{-1} permitted to be applied to SRWC in Sweden, although far lower than that in other countries, might nevertheless be high for sustainable use, if the needs of SRWC, *i.e.* *ca.* 10 kg P ha^{-1} yr^{-1}, were taken into account (see theoretical calculations above). Minimisation of the amount of P supplied with sludge–ash mixtures would imply fewer risks for contamination due to P losses. In that case, the amounts of heavy metals applied would also simultaneously decrease, and sustainability in terms of heavy metals would consequently also improve. In recent years, wastewater plant operators have shown increased interest in disposing of sludge in SRWC. This is mainly a consequence of new EU laws, which do not permit sludge disposal in landfills from year 2005 onwards; thus an increase in the area of SRWC in Sweden is a possible future scenario. It is doubtful, however, whether the interests of wastewater operators are compatible with any minimisation of the P supply with sludge–ash applications to SRWC. In any case, the fate of P in the soil, not only after sludge–ash applications, but also after irrigation with wastewater types that supply large amounts of P to the soil (*e.g.* irrigation with log-yard runoff), and the effects on environmental quality, should be more closely studied. This is a prerequisite for the wider application of municipal or industrial residues to SRWC in future.

Conclusions

The main conclusions drawn in this thesis concerning the sustainable use of the various residues applied to SRWC—which implies minimal environmental hazards and good SRWC growth—are:

- N-leaching to the groundwater is a potential threat when high N loads are applied to SRWC fields by high wastewater irrigation rates;
- Comparison of experimental data from full-scale SRWC fields irrigated with wastewater in central Sweden suggests that the N load that can be treated is much higher than the N requirements of SRWC, and depends on site-specific conditions;
- When the concentration of hazardous compounds in the wastewaters is low—as in log-yard runoff—the phytoremedial efficiency of SRWC is satisfactory and is based on irrigation intensity;
- When hazardous compounds are present in the wastewater—as in landfill leachate, which contains high concentrations of salts and of organic compounds—the irrigation rate should be adjusted to avoid growth reduction;
- Genetic differences in salt tolerance and growth performance between willow clones call for careful selection of clones to suit specific situations;
- Leaf length can be used as a rapid diagnostic tool for identifying stress, in fields irrigated with landfill leachate, to permit adjustment of the irrigation rate, hence to avoid growth reduction;
- Fertilisation of SRWC with legally permitted sludge–ash mixtures does not pose a substantial threat to sustainability, in terms of heavy metals;
- When SRWC is fertilised with sludge–ash, a reduction in the total Cd amounts in the soil is expected to occur after harvest.

In addition, when irrigation of SRWC with different wastewaters is practised, any decision concerning the irrigation regime must take into account climatic factors (such as precipitation, length of growing season), soil type, residue composition, and species or clone characteristics (evapotranspiration rate, climatic adaptability, pest tolerance), if both a high growth rate of SRWC and sustainability are to be attained.

Furthermore, when sludge–ash mixtures are applied to SRWC, better management practices, *e.g.* the introduction of suitable clones in terms of metal uptake, the selection of higher-yielding clones, and shorter harvest intervals, would contribute to better sustainability in terms of heavy metals. However, the high P loads applied in a single application to SRWC may constitute a future hazard to the environment.

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