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Design of a zero liquid discharge leachate treatment system using an evapotranspiration willow bed

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ABSTRACT

While zero liquid discharge (ZLD) wetlands have been successfully used for domestic wastewater treatment, adapting this technology to treat other wastewaters such as leachate could be very attractive for some industries concerned with meeting increasingly stringent environmental regulations. Leachate treatment typically implies large volume of water that are entirely dependent on rainfall and therefore highly variable both throughout the year and between years. Current design guidelines for zero discharge willow systems limit system flexibility because they are based on rough theoretical estimates of evapotranspiration. This discuss the applicability of ZLD treatment through a willow bed evapotranspiration (ET) applied to the treatment of industrial leachate that has high and variable hydraulic loading rate and low contaminant and salt concentration. We propose a base design and, through detailed and long-term hydrological modeling of such a treatment system, investigate how various design and management decisions can affect sizing, efficiency, and overall feasibility of the technology. We showed that considering ET optimization factors (e.g. fertilization and organic substrate) was essential for ZLD to be achieved over a 20-year period in northern continental humid climate and that the ratio between cumulative annual ET of the willow bed and cumulative annual rainfall should be at least 1.5. When varying the leachate collection area, it was found that a ratio of willow bed area to collection area between 0.5 and 0.7 should be expected for an optimized design in this specific climate, were land area and storage volume remain the most limiting factors. Regarding storage volume, several management options can be applied to reduce the volume of storage required. We also highlight that a risk attenuation strategy should always be included in the design of a ZLD wetland system. Our study suggests that ZLD wetlands constitute a green technology that represents a serious alternative treatment method for pretreated leachate, while offering many benefits such as low maintenance and energy costs, valorization of contaminants such as nitrogen or phosphorus through biomass production, and, most importantly, zero contaminant discharge to the environment. Finally, we propose future research opportunities and other possible applications for further development of the technology.

1. Introduction

Every year, industries must treat a large, variable volume of water, due to rainfall leaching through various wastes or products (e.g. landfills, mine wastes, stored treated wood poles). Among the available solutions used for industrial wastewater treatment, some aim to reduce the volume of water released into the environment to zero and are referred to as zero liquid discharge (ZLD) systems. Such systems were first developed to allow different industrial sectors to reduce both water consumption and treatment costs (Koppol et al., 2004). This type of approach can also allow industries to avoid having to obtain a discharge permit for their contaminated effluent. The concept of ZLD is used increasingly to address a number of the different constraints and difficulties associated with wastewater treatment (Tong and Elimelech, 2016). Typical ZLD systems can take various forms, the simplest being evaporation ponds where wastewater is stored until it passively evaporates. Not only does this type of system fail to enable reuse of the water, it also requires a very large area and is feasible in very few contexts, such as arid and semi-arid climates. In contrast, some highly sophisticated systems combine physical (e.g. water purification through energy-fueled evaporators) and chemical (e.g. adding chemicals to precipitate a specific compound) steps to both purify water and raw materials that can be

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reused. These latter systems can be very efficient but are typically energy demanding with high installation and operation costs.

Phytotechnology-based ZLD systems were introduced in the 1990s in the form of constructed wetlands planted with willows. They were used to treat domestic wastewater in Denmark and designed to generate zero effluent (Gregersen and Brix, 2001; Brix and Arias, 2005, 2011). Since then, this technology has also been used in Ireland (O'Hogain et al., 2011; Curneen and Gill, 2016), and feasibility studies were successfully carried out in Mongolia (Khurelbaatar et al., 2017). Such constructed wetlands are now referred to as willow wetlands, willow systems, willow beds or zero-discharge wetlands (Kadlec and Wallace, 2008; Dotro et al., 2017). The targeted mechanism for reducing the volume of the effluent is evapotranspiration (ET), in which the system loses water by evaporation and plant transpiration. This natural process is sometimes utilized in environmental engineering, especially to treat contaminated landfill leachate (Białowiec et al., 2011). Using this form of green technology in industrial wastewater treatment offers a number of advantages over conventional approaches, including low maintenance and energy costs, concentration of the contaminants or by-products in a confined compartment (i.e. the wetland substrate), valorization of contaminants such as nitrogen or phosphorus through biomass production, and zero discharge to the environment.

Willow systems for domestic wastewater treatment are typically sized according to hydraulic and surface loading rates for population equivalent values (Dotro et al., 2017), and current design guidelines for willow systems are often based on rough estimations of ET (e.g. mean annual crop coefficient of 2.5 times a reference ET; Brix and Arias, 2005). Furthermore, domestic wastewater represents a relatively constant and predictable hydraulic loading and low phytotoxic potential. To our knowledge, no design recommendations or criteria have been proposed for the treatment of other types of wastewater or for more complex treatment systems. Industrial leachates typically have very high and variable hydraulic loading rate and may have high contaminant and/or salt concentrations, which would require specific consideration in the design process or even compromise reduce ZLD feasibility if used in their raw form (e.g. leachates from young landfill cells or acid mining drainage; Brennan et al., 2016, Simate and Ndlovu, 2014). Furthermore, the discharge regulations are not the same for all types of leachates, which would also influence the decision of using a ZLD approach and the system management.

In this study, we focus on the treatment of industrial leachate that (1) has high and variable hydraulic loading rate, (2) is either pre-treated or that contain low contaminant and salt concentration, and (3) for which discharge regulations are problematic and justify the interest in a ZLD solution. Our objective is to propose a design for a flexible and durable ZLD system incorporating an evapotranspiration willow wetland that is hydraulically limited but could also be applied to related leachate characteristics. Through hydrological modeling of such a treatment system, we also investigate how various design aspects could be used to significantly optimize management practices. Our modeling approach was based on a detailed analysis of ET temporal variation and water flow management.

2. Materials and method

2.1. Model description

2.1.1. Model development

The design of a ZLD for treating leachates is highly dependent on climatic regime. The surface required for zero discharge of a given leachate collection area would be minimal under a tropical, drier climate with highly predictable precipitation. Our model was developed and tested under the more stringent conditions of the humid temperate continental climate of southeastern Canada (Québec). This region has long, freezing winters and warm, humid summers. Precipitation levels are relatively high (annual average around 1 m, 20% falling as snow) and occurring all year round, with occasional dry spells, especially in summer. High evapotranspiration only occurs during the relatively short growing season. Temporary leachate storage may thus be necessary for winter precipitation and extreme rainfall events, and to provide sufficient irrigation to plants during dry summer periods.

ZLD evaporative efficiency also depends on the water composition of the leachate. As mentioned in the introduction, pretreatment would be recommended for highly contaminated leachate to prevent willow toxicity and rapid accumulation of contaminants and salts in the ZLD bed, and to expand its overall lifespan. Furthermore, since willow biomass production is strongly correlated to ET (Martin and Stephens, 2006) and generally increase with nutrient availability (particularly nitrogen; Fabio and Smart, 2018), fertilization may be necessary to maximize the willow bed ET when treating nutrient-deficient leachate.

The system design modelled here comprises an open tank or pond (hereafter referred to as an open collection tank) that stores wastewater or collects leachate seeping out from contaminated materials; being open, the collection tank receives rainfall as well (Fig. 1). A pre-treatment step is included - here, in the form of a treatment wetland (physical, chemical and biological removal processes) - to ensure that the outflow of this compartment and that is used to irrigate the evapotranspiration willow bed has a low contaminant charge (Fig. 1). We assume that gross filtration and settling of large particles occur prior to the pre-treatment step. Finally, two equalization tanks (equalizing tank 1 and equalizing tank 2), are connected to the collection tank and the willow bed, respectively, to manage water flowrate variability in the system. Although the two equalization tanks could have been combined into one, their separation ensures that only pre-treated water enters the willow bed, thus increasing its life span, and allows treated water to be separated from partially treated or raw wastewater. As illustrated in Fig. 1, the only water input in the system is the rainfall occurring over the collection tank, the treatment wetland and the willow bed, and the output occurs through evaporation (E) from the collection tank and evapotranspiration (ET) from the treatment wetland and the willow bed. The input water not lost through evapotranspiration moves from one compartment to the other and the closed circuit enables operation of the system with ZLD. The model was developed to produce a daily computation of the value of every water flux and the volume of water contained in each component of the system after all water exchanges have occurred, based on the classical hydrological balance equations (see supplementary material for detailed equations used).

To maintain the general water flow described above, a set of management rules was implemented in the model (see Supplementary material). Globally, these rules ensure that (1) water is always available for E and ET in both the collection tank and the willow bed and that sufficient water is available in the collection tank for irrigating the treatment wetland at all times, (2) overflow of the collection tank, the treatment wetland and the willow bed are conveyed to the designated compartments (see Fig. 1), (3) the water level in the collection tank, the treatment wetland and the willow bed is lowered before winter to prevent root and pipe damage in the treatment wetland and the willow bed due to water freezing and to prevent spring overflow in the collection tank, and (4) if, as a last-resort remedy, water must be pumped out of the system or discharged to the environment, the effluent will have gone through both primary treatment in the regular treatment wetland and secondary treatment in the willow bed, and therefore contain the lowest concentration of contaminants possible.

2.1.2. Model parameterization and calibration

Based on the model design presented in Section 2.1.1, we can determine five categories of parameters that are needed for the model to operate: component design, water flux management, plant parameters, evapotranspiration and meteorology (see Supplementary material). Some of the parameters, such as meteorological data, are external and cannot be controlled, while auxiliary parameters, like component design, are determined by the user. Other parameters such as

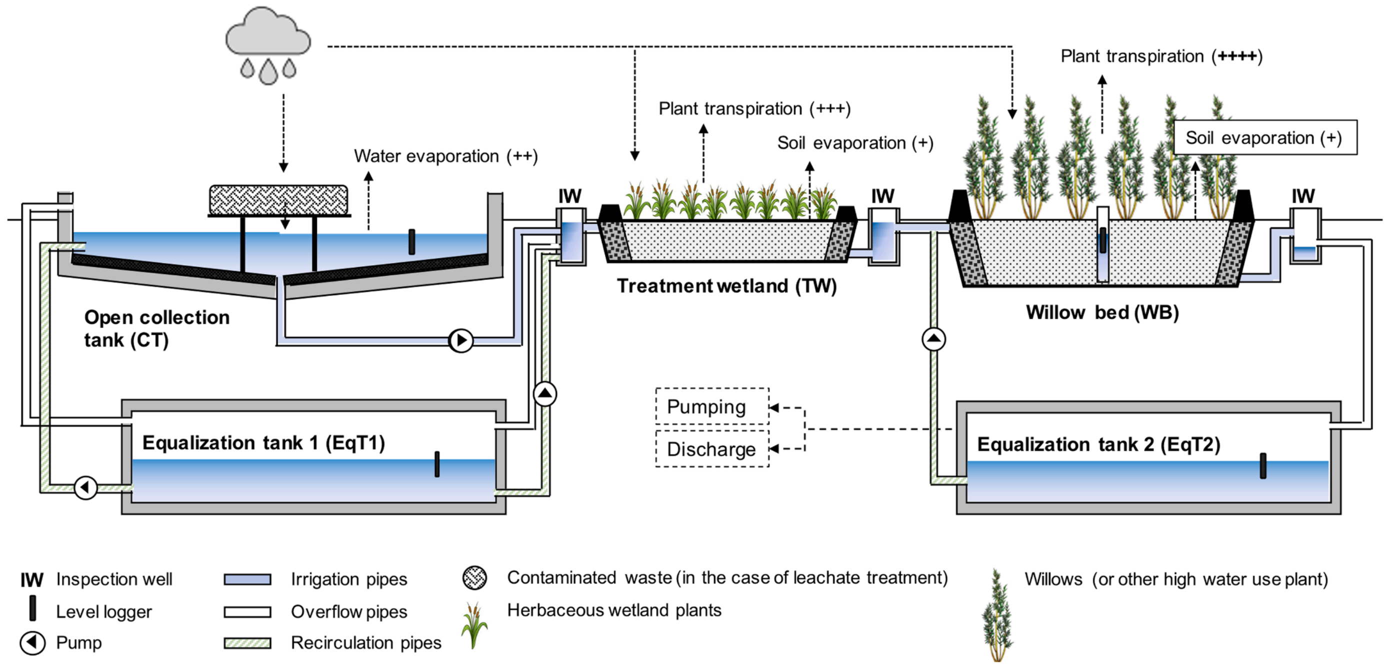

Fig. 1. Design proposed to operate a leachate treatment system with zero liquid discharge using an evapotranspiration willow bed. The drawing is not to scale. Water input to the system is exclusively through rainfall on the system compartments, and water output is through evaporation and transpiration at relative magnitudes indicated by the number of "+" signs. Pumping or discharge of water is also possible from the second equalization tank. An external reliable source of water may be added for occasional plant irrigation during exceptional dry spells.

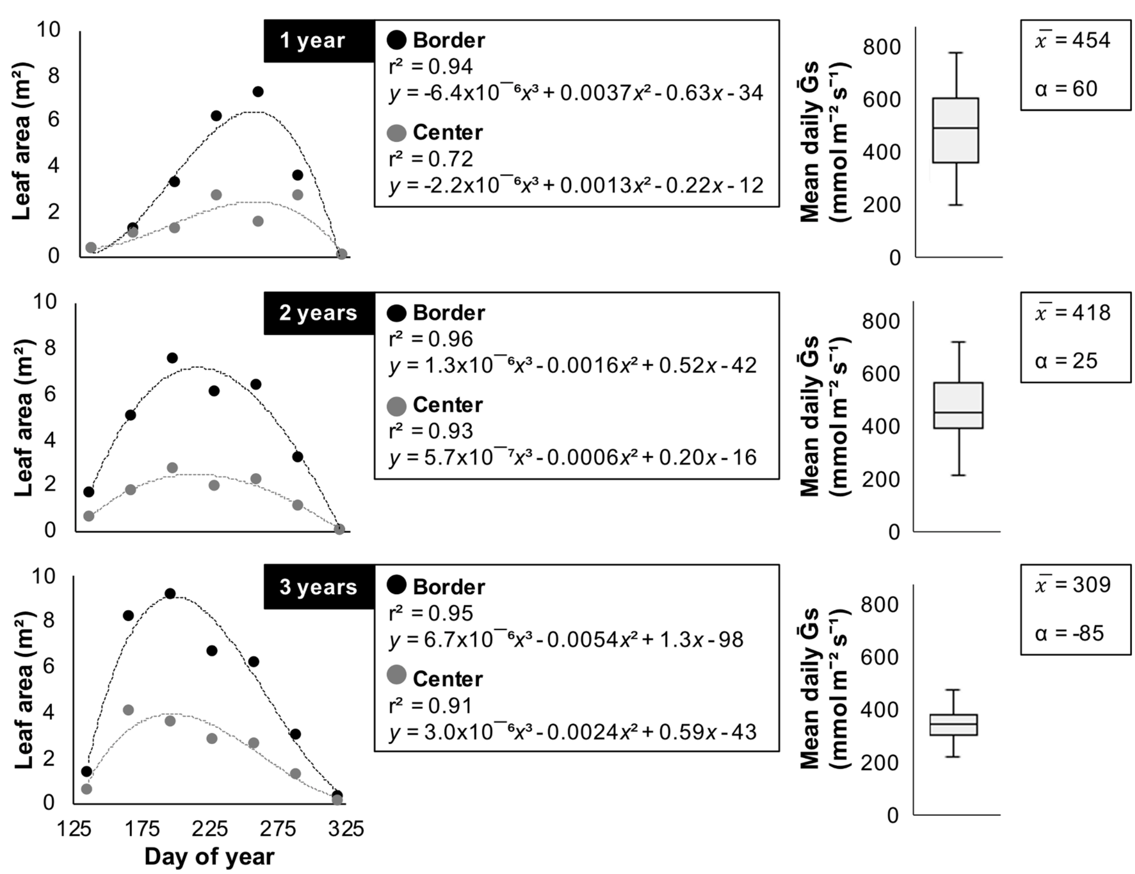

Fig. 2. Mean leaf area per individual willow, according to stool age, position in the wetland and time of year, and mean daily general stomatal conductance ($\bar{G}_s$) according to stool age. ($\bar{G}_s$) average ($\bar{x}$) annual value and its deviation from the 3-year mean value (α) are presented.

evapotranspiration are external but can be modulated through management decisions (see *Design optimization opportunities*, Section 2.3). ET calculations are based on a previous study conducted on a willow bed planted with *Salix miyabeana* 'SX67' (Frédette et al., 2019a; see Supplementary material) and leaf area index (LAI) calculated on site (Fig. 2). Evaporation (E) from the collection tank is estimated to be about 80% of the Penman-Monteith reference evapotranspiration (ET_0; Allen et al., 1998), treatment wetland evapotranspiration (ET_{TW}) is estimated to be equal to ET_0 (crop coefficient, or K_{et}, of 1).

2.1.3. Model validation

This modeling study uses data collected from a demonstration scale leachate treatment system set up and operating in Québec, Canada since 2012 as a reference. This system was built to evaluate the performance of different wetland processes for treating leachate collected on a storage site for treated wood poles; it was not designed as a ZLD and its effluent is discharged into a municipal sewer. A general description of the system and its design parameters is provided in the Supplementary materials. For the purposes of this study, the following parameters were monitored on the reference site during the 2016, 2017 and 2018 operation seasons: the volume of water pumped into the treatment wetlands (in*TW*; m^3), the volume of both willow bed affluent and effluent (*inWB* and *outWB*; m^3) and willow bed evapotranspiration (*ETWB;* m^3). To validate our model, we slightly modified the conceptual model presented in Section 2.1.1 so that the overflow of the willow bed would flow out of the treatment system instead, and both equalization tanks were removed, since there is currently no such tank on site. We simulated only the operating seasons, which were from May 9 to November 30 in 2016 (206 days), from May 15 to November 9 in 2017 (179 days) and from May 10 to October 31 in 2018 (175 days). Following the simulations, we were able to calculate *inTW, inWB, outWB, ETWB*. We then plotted observed values against predicted values and calculated the data determination coefficient (R^2) compared to a prefect correlation model output ($x = y; R^2 = 1$) to assess the model prediction capacity (Piniero et al., 2008).

2.2. Evapotranspiration optimization

Although evapotranspiration is mainly driven by climate and plant physiological traits, providing favorable growing conditions can enhance plant transpiration. For willows in particular, a first way of promoting ET is to provide a constant water supply (Frédette et al., 2019b). In this model, we fixed a threshold water level (15 cm below ground) under which additional irrigation is provided to the willow bed, to ensure that water availability to willows is always maximal.

2.2.1. Wetland aspect ratio

Varying the aspect ratio (L:W, length over width) of the willow bed, for a given surface area, could also represent an opportunity for optimization, by increasing ET per unit surface. The higher the aspect ratio, the longer the perimeter of the willow bed, with willows growing on the perimeter of the bed having a significantly higher LAI (up to 300% more than those growing in the center; Fig. 2), which is directly correlated with ET (Frédette et al., 2019a). To test the effect of the L:W variation on sizing criteria, we simulated the operation of the treatment system using a regular shape ($L{:}W = 1.5$) and then an elongated shape ($L{:}W = 10$). The LAI was adjusted directly in the model according to the number of willows growing on the border (W_{border}) and in the center of the bed (W_{center}). The methods used to calculate LAI, W_{border} and W_{center} are described in Supplementary material.

2.2.2. Fertilization

Another way of enhancing willow ET is to increase the supply of nutrients available for plant growth (i.e. fertilization). In two studies where only the fertilization amount varied between treatments, it was demonstrated that fertilized willows (*S. alba*) lost 96% more water through evapotranspiration than unfertilized ones, and that increasing the level of fertilization could increase ET by another 51% (Guidi et al., 2008; Pistocchi et al., 2009). Considering that the ET model used was calibrated using slightly fertilized willows (Frédette et al., 2019a), we applied an ET increase coefficient (αF) of 1.51 to simulate the effect of high fertilization on the sizing criteria.

2.2.3. Substrate characteristics

Finally, we wanted to simulate the effect of different substrates: (1) a sand substrate, that provides good drainage, and (2) an organic substrate that provides increased organic matter and water retention in the root zone but is more susceptible to compaction and clogging. In a mesocosm study, we reported that ET of willows grown in sand achieved about 77% of the ET of those grown in a coconut fiber substrate and about 65% of the ET of those grown in a highly organic potting substrate (Frédette et al., 2019c). The ET equations used in our model were calibrated for willows grown in a peat and sand substrate (Frédette et al., 2019a). Although peat and coconut fiber substrates are comparable because they share similar physical properties, Bañón et al. (2009) reported a 23% increase of ET in peat versus coconut fiber. Therefore, in our model, ET calculated according to the method described by Frédette et al. (2019a) is considered optimized in terms of substrate and an ET decrease coefficient (αS) of 0.65 was used to simulate the effect of using a sand substrate.

2.2.4. Coppicing cycle

Using a willow planted wetland implies that the woody biomass must be coppiced on a 2- to 4-year coppicing cycle, as is often suggested for willow plantations designed for biomass production (Bullard et al., 2002). Coppicing is also essential to maintain a high level of plant activity, which is correlated to a high evapotranspiration rate (Dotro et al., 2017). However, recently coppiced willows have less leaf area available for transpiration compared to mature trees. Based on the data collected at the reference site, we found that average LA typically increased with the age of a stool - root or stump of the shrub from which shoots spring after coppicing - and that, inversely, average $\bar{G}_s$ decreased with stool age, average value being maximal for stools of one year and significantly lower for stools of three years (Fig. 2). To minimize the effect of coppicing on evapotranspiration, alternately coppicing different sections of the willow bed has been suggested (Gregersen and Brix, 2001; e. g. one half of the bed coppiced one year, and the other half coppiced the following year, for a 2-year cycle). For the purpose of this modeling study, it was assumed that a 2-year coppicing cycle was used, so that, every year, half of the willow stools were 1 year old, and the other half were 2 years old.

2.3. Simulation scenarios

2.3.1. Simulation plan and design optimization

We simulated a time frame of 20 years of operation, which we considered appropriate to represent a wide range of meteorological variations, particularly rainfall. The first objective of the simulations was to determine how the general design proposed in Section 2.1.1 performed in managing the varying volumes of leachate generated from rainfall at the reference site. Then, various combinations of optimization options (see Section 2.2 and Fig. 3a) allowed us to assess the impact of management decisions on system performance. We thus simulated each design parameter combination by setting the willow bed area to 2000 m^2 to compare performance in terms of storage volume required, ET volume and overflow frequency. A meteorological database covering 20 years (1996 to 2015) and including all the necessary parameters to calculate ET_0 and ET_{wb} was created with data from Environment and Climate Change Canada (ECCC) (2019).

2.3.2. Determination of sizing criteria

The relationships obtained were then implemented in the model so

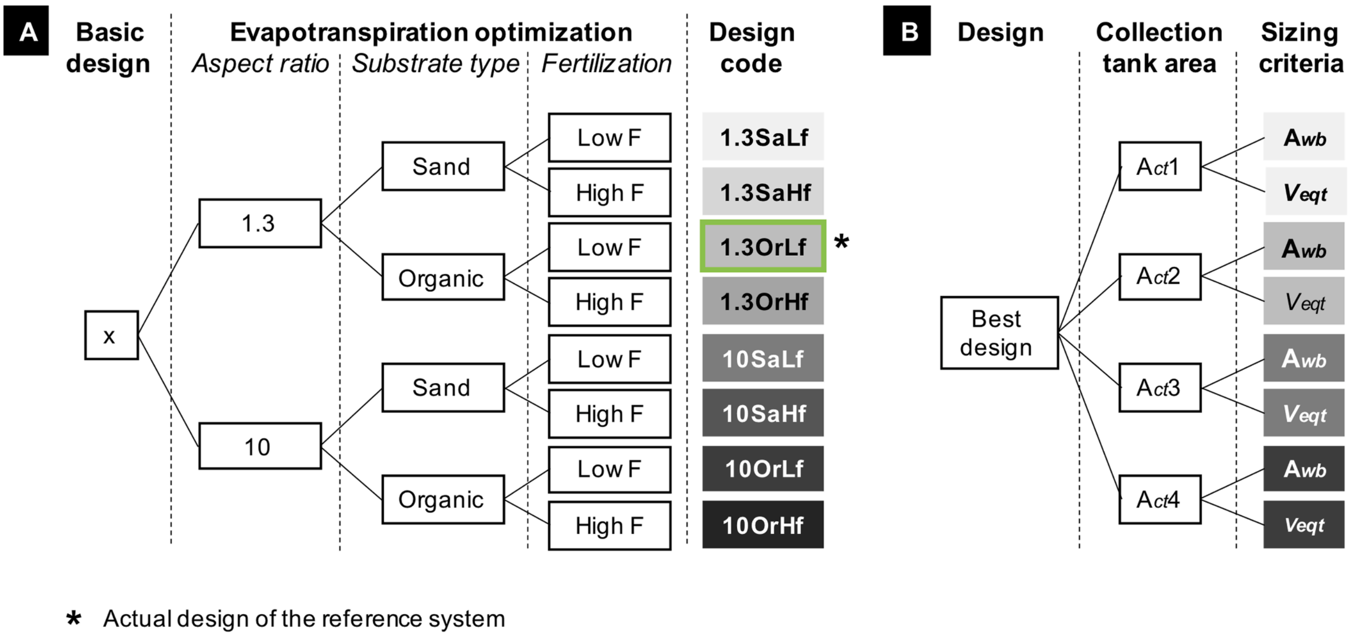

Fig. 3. **A.** Simulation plan used to model the operation of a ZLD treatment system using an evapotranspiration willow bed and determine the best design. **B.** Simulation plan used to establish a relationship between the area of an open leachate collection tank and the willow bed area required to achieve a ZLD.

that when changing the collection tank or the willow bed area, the corresponding equalization tank volume required was automatically adjusted. Once the best design parameter combination was identified (see Section 2.3.1), the willow bed was sized by increasing its area until the simulation results made it possible to achieve a ZLD over a 20-year period of simulation. We also simulated the operation of the system with different areas of the leachate collection tank to determine a numerical relationship between the collection tank area, the willow bed and the equalization tank sizing criteria (Fig. 3b). Such a relationship could be helpful for future use of ZLD wetlands, for example in assessing the feasibility of accommodating the technology in the available space.

3. Results

3.1. Model validation

Based on meteorological data from 2016, 2017 and 2018 and the design parameters of the reference site, we were able to model several components of the hydrological balance of the reference site treatment system with a determination coefficient (R^2) of 0.68 (Fig. 4). Both the influent and the effluent of the willow bed tend to be overestimated by the model (25 and 37% respectively), which can be the result of underestimating ET_{TW} and ET_{WB}; ET_{WB} appeared to be underestimated by the model (20%; Fig. 4). We therefore concluded that the model is conservative, which could slightly increase the risk of over-dimensioning the system compartments. Because the treatment system modelled is intended to reduce the risk of discharge by overflow, we consider a conservative model to be appropriate and the validation results satisfactory.

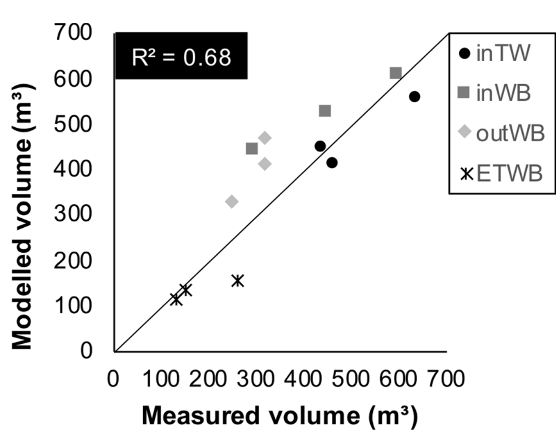

Fig. 4. Comparison of modelled and measured values of some components of the hydrological cycle of a water treatment system using a treatment wetland and a willow bed. inTW = treatment wetland affluent, inWB = willow bed affluent, outWB = willow bed effluent, ETWB = evapotranspiration of the willow bed.

3.2. Design optimization and sizing criteria

When performing the 20-year simulation with a fixed the willow bed area of 2000 m^2, only the designs optimized in terms of substrate and fertilization (*1.3OrHf, 10OrHf*) led to ZLD (Table 1). This result coupled with the calculated ratios between cumulative annual ET of the willow bed and cumulative annual rainfall suggests that cumulative annual ET need to be at least 1.5 times higher than annual rainfall for ZLD to be achieved over a 20-year period in northern continental humid climate. Conversely, designs with low fertilization and sand substrate (*1.3SaLf, 10SaLf*) were the least preforming, with overflow occurring at least once per year, 19 of the 20 years simulated and showing the highest mean yearly cumulative OF (1480 to 1540 m^3/a). When at least one optimization option was added to the design (*1.3SaHf, 1.3OrLf, 10SaHf, 10OrLf*), in comparison with the actual design of 1.3OrLf, yearly cumulative OF was reduced (830 to 990 m^3/a) but overflow frequency remained high, from 17 to 18 years out of 20 years. Overall, designs with a 10:1 aspect ratio required less storage volume (15% to 25%) and generated less yearly cumulative OF (4% to 14%) than their homologous designs with a 1.3:1 ratio, except for the best performing design, *1.3OrHf* and *10OrHf*, where the 10:1 aspect ratio required 360 m^3 more storage volume. Furthermore, mean yearly cumulative ET was 7% lower with the *10OrHf* design than with *1.3OrHf*. When looking at the fate of water in the system over the course of the 20 years with the design including all the optimization options (*10OrHf*), we found that for 13 of the 20 years, the willow bed was completely emptied (no ET possible) at least once during the summer months, a situation that occurred only 3 years

Table 1
Results of a 20-year simulation (1995–2015) of the complete operation of a zero liquid discharge leachate treatment system using an evapotranspiration willow bed. Values in bold (design 1.3OrLf) represent the actual design of the reference site. In the design codes, 1.3 and 10 represent the willow bed aspect ratio (length:width), Sa and Or the soil type (sand or organic, respectively) and Lf and Hf the fertilization level (low or high, respectively).

Design code	ET_{wb} (m^3/a)	ET/R	OF (m^3/a)	YWO (a/20a)	V_{eqt2} (m^3)	EqT2 sizing equation
1.3SaLf	1100 ± 150	0.74 ± 0.2	1540 ± 620	19	1990	$1.1[0.82A_{WB} + 210 - (Vmax_{WB} - Vmin_{WB})]$
1.3SaHf	1630 ± 210	1.2 ± 0.2	990 ± 680	18	2010	$1.1[0.72A_{WB} + 520 - (Vmax_{WB} - Vmin_{WB})]$
1.3OrLf	1690 ± 220	1.2 ± 0.2	940 ± 670	18	1630	$1.1[0.68A_{WB} + 270 - (Vmax_{WB} - Vmin_{WB})]$
1.3OrHf	2390 ± 430	1.6 ± 0.3	0	0	2480	$1.1[1.2A_{WB} + 1.7 - (Vmax_{WB} - Vmin_{WB})]$
10SaLf	1180 ± 160	0.81 ± 0.2	1480 ± 590	19	1560	$1.1[0.61A_{WB} + 360 - (Vmax_{WB} - Vmin_{WB})]$
10SaHf	1770 ± 240	1.2 ± 0.3	850 ± 640	17	1700	$1.1[0.71A_{WB} + 280 - (Vmax_{WB} - Vmin_{WB})]$
10OrLf	1810 ± 240	1.2 ± 0.3	830 ± 660	18	1230	$1.1[0.72A_{WB} + 280 - (Vmax_{WB} - Vmin_{WB})]$
10OrHf	2220 ± 490	1.5 ± 0.3	0	0	2840	$1.1[1.3A_{WB} + 130 - (Vmax_{WB} - Vmin_{WB})]$

ETwb = evapotranspiration of the willow bed, ET/*R* = ratio between cumulative annual willow bed ET (mm/a/m^2) and cumulative annual rainfall (mm/a/m^2), OF = overflow (volume of water being discharged or pumped out of the system), YWO = years without overflow, Veqt2 = volume required for the second equalization tank.

out of 20 when using the *1.3OrHf* design. Water storage needs increase when ET is increased, because, as the maximum ET increases, so does the gap between minimal and maximal cumulative net inflow (which is used for storage tank sizing).

For the first simulations in which the collection tank area was set at 2240 m^2 (actual size at the reference site), we found that a volume of 1910 m^3 was required for equalization tanks 1. For each willow bed design tested, we were able to establish a linear relationship between the area of the willow bed and the volume of equalization tanks 2 needed (Table 1). In the end, when using the *1.3OrHf* design, we found that 1750 m^2 of willow bed and 2170 m^3 of second equalization tanks were required for ZLD to be attained within a 20-year time frame, compared to 1590 m^2 of willow bed and 2280 m^3 of second equalization tank when using the *10OrHf* design. Choosing which of the two would be the best performing design therefore depends on user preferences and limitations. Our results showed a linear relationship between the willow bed area required and the collection tank area (Fig. 5), with ratio A_{WB}:A_{CT} of about 0.7 for the *1.3OrHf* design and 0.5 for the *10OrHf* design.

3.3. Global system performance

Several observations can be made based on tracking water as it circulates through the system compartments of the best designs over the period of 20 years simulated (Fig. 6). Regarding the willow bed, we can see that the minimal level could not be maintained in some years (particularly 1996, 2001, 2002 and 2003 for *1.3OrHf* and 2002 and 2003 for *10OrHf*) because both equalizing tanks had been emptied during the preceding years. Wetland irrigation might have been necessary during those years. From summer 2004 to autumn 2014, there was a continuous increase in the use of equalizing tank 2, which reached its maximal capacity in 2011, 2012, 2013 and 2014 for *1.3OrHf*, and in 2013 and 2014 for *10OrHf*. By the end of the simulation, the volume of water in equalizing tanks 2 was lowered, but the trend suggests that it might be necessary to discharge a certain volume from this compartment to continue to operate the system beyond 20 years. This type of exceptional discharge should then be managed according to applicable regulations, since contaminants could accumulate in the willow bed substrate and lead to an effluent concentration requiring transport to a conventional or special wastewater treatment facility. The collection tank was systematically filled up during winter and emptied during the following summer. Much like equalizing tank 2, equalizing tank 1 was used continuously after several years of operation (from winter 2006 to the end of the simulation). However, while equalizing tank 1 maximal value was reached once in 2014 for *1.3OrHf*, it never reached more than about 65% of its volume for *10OrHf*. The methodology used for equalizing tank 1 sizing considers only water coming in from the overflow of the collection tank (which is predictable), and not the water going out of equalizing tank 1 to irrigate the treatment wetlands when water levels in the willow bed and equalizing tank 2 are too low (less predictable), which could explain oversizing of equalizing tank 1.

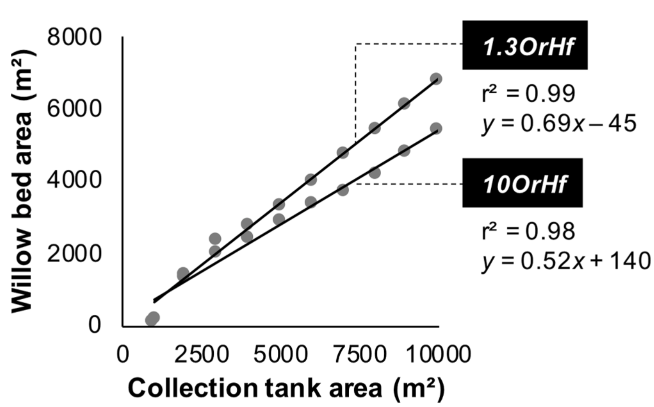

Fig. 5. Relationship between the leachate collecting tank area and the willow bed area required to achieve a zero liquid discharge effluent, when the design is optimized in terms of substrate (organic, Or, versus sand, Sa) and fertilization (high, Hf, versus low, Lf) and for two different aspect ratios (1.3 and 10).

4. Discussion

Variable flows and evapotranspiration potentials represent the most challenging aspects in using ZLD willow beds to treat industrial leachates. Our model was able to predict water flows in a reference treatment system with satisfactory results. We were able to establish a numerical relationship between the area of an open collection tank and the surface area of willow bed required for a system to attain ZLD, which could help guide design process in the future. Analysis of ET-related plant parameters ($\bar{G}s$ and LAI) at the reference site highlighted their temporal variation and lead to suggest that a two-year coppicing cycle should be preferred over a 3-year cycle to maintain maximal ET. Modeling the daily operation of a complete system over a 20-year period showed that attaining a ZLD is feasible in a humid continental northern climate, presuming that sufficient land area and storage volume are available. It also highlighted several aspects to consider in design and management decisions. For example, modulating ET through substrate selection and fertilization can significantly reduce the storage volume and willow bed area needed; sizing the willow bed with a high aspect ratio can further optimize the system, but to a lesser extent.

4.1. Feasibility of the technology

In this study, the feasibility was assessed only for the hydraulic perspective, by iteratively increasing compartments size until zero-liquid discharge was achieved. However, the only design scenarios where ZLD could be systematically achieved over a 20-year period were

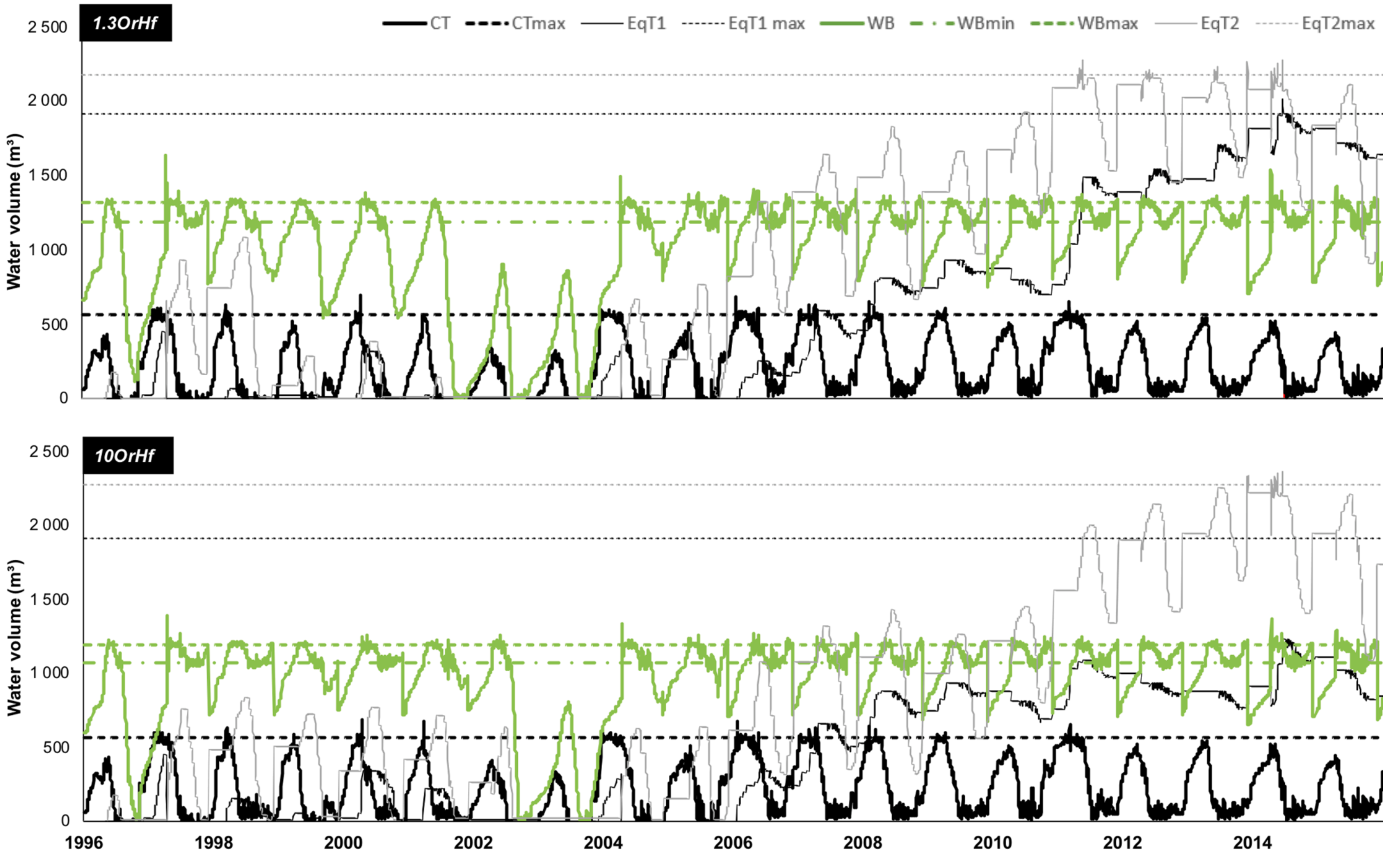

Fig. 6. Daily variations, simulated for 20 years of operation, of the water volume in the different compartments of a ZLD treatment system using an evapotranspiration willow bed as a tertiary treatment. Results are presented for two different aspect ratios (1.3 and 10) and for highly fertilized willows growing in an organic substrate. The volume of water in the upstream secondary treatment wetland (V_{max} = 26 m^3) is not shown.

those including at least two ET optimization option, and those in which the ratio between annual ET and rainfall was equal or greater to 1.5. Theoretically speaking, ET equal to rainfall (ratio of 1) should be sufficient to achieve ZLD, confirming that managing high hydraulic loading variability over a long period requires a substantial buffer (50% in this case).

Although not related to technical feasibility *per se*, other considerations could influence the suitability of the technology for a given case scenario, such as available land area and financial and/or human resources, leachate composition, available risk tolerance, etc. As mentioned, this article refers to a treatment system that is limited by its hydraulic capacity rather than its pollutant loading capacity, either because of pre-treatment or a low contaminant concentration of the leachate, meaning that salts and/or contaminant accumulation in the substrate or system clogging are not major issues. Table 2 presents several factors to consider when assessing the willow bed approach compared to other technologies that lead to zero contaminant discharge.

Table 2
General comparison of four treatment options that enable achievement of zero discharge of contaminated water.

	Willow bed	Evaporation pond	Thermal processes	Reverse osmosis
Capitalization costs	++	+	+++	+++
Operational costs	+	+	+++	++
Resource consumption	+	+	+++	++
Water reuse	P	N	Y	Y
Solids recovery	N	N	Y	Y
Land area required	++	+++	+	+
Residual wastes[1]	+	++	+	+

Y = yes, *N* = no and *P* = potentially.
[1] Contaminant concentrates (thermal processes and reverse osmosis) or contaminated substrate (willow bed and evaporation pond).

Potential for increasing technical feasibility, adapting the approach to other climates or wastewaters and other limitations will be discussed in the following sections.

4.2. Evapotranspiration optimization

Our results, along with those of previously published studies, confirm the relevance of using willows in phytotechnology-based ZLD systems. *Salix* species have shown a general tendency to increase ET when water supply is not a limiting factor (Frédette et al., 2019b), and their suitability for coppicing ensures that plant growth and ET rates are maximized. While the effect of willow stools and root age on its productivity and biomass production has been studied (Mola-Yudego and Aronsson, 2008; Volk et al., 2011), little attention has been given to its effects on ET. The few studies comparing ET of willows of different ages generally compared the establishment year to subsequent years (Gregersen and Brix, 2001, Białowiec et al., 2011; Guidi et al., 2008). However, ET through the establishment year is expected to be lower, and such information does not provide information on how ET is affected by coppicing when the root system is well-established. Our method and results are an important step forward in assessing how stool age affects the volume of ET produced by a well-established fast-growing willow shrub. However, our results might be species specific and would benefit from further field validation.

While it is well known that fertilization can increase ET by increasing general plant growth and activity, determining an increase coefficient that can be used in an ET predictive model is complex since many factors are susceptible to influencing this coefficient, like species nutrient needs, species maximal potential ET rate, composition of the fertilizer used, concentration applied and water availability. Deriving an ET increase coefficient from a study testing both fertilized and unfertilized willows in otherwise exact growing conditions still allowed us to highlight that fertilization may be the most important factor to optimize an evapotranspiration system. The study used for reference (Frédette et al., 2019c) had the advantage of using the same species as the reference site, but also the limitation of being a lysimeter and green house experiment, the results of which could not translate perfectly to field conditions.

Therefore, although we strongly recommend considering the soil type and fertilization influence on ET when designing an evapotranspiration willow bed, determining more precise coefficients for these two factors (for example, by conducting a pot experiment with the species, considering the nutrient sources and substrates to be used) could be necessary before implementing a full-scale system.

4.3. Management practices and other design considerations

It seems that even if the collection tank was completely emptied during the summer months, the volume of this compartment was not sufficient to accommodate the cumulative rainfall of late autumn, winter and spring. Increasing the depth of the collection tank without expanding its surface area, could help prevent off-season overflow of the collection tank, and further reduce the volume of equalizing tank 1 required. While this highlights the importance of considering ET and rainfall variability over time, some nuances should be pointed out. First, in our study, equalizing tank 1 was sized based on the actual dimensions of the collection tank on the reference site (2240 m^2, 543 m^3). Constructing a collection tank or pond with a greater volume (i.e. increasing the depth) would substantially reduce the volume of storage needed to equalize this compartment, while also preventing spring overflow. As discussed in Section 2.1.1, the two equalizing tanks could also be combined into a single tank for the whole system, which would also significantly reduce the volume of storage required. However, this would mean that potential discharge of the system (e.g. for system maintenance or following an extreme weather event) would release a mix of treated and raw wastewater that would generate further treatment costs. Treated wastewater alone (from equalizing tank 2 in the two equalizing tanks scenario) could potentially be discharged in compliance with environmental regulations.

It is also important to keep in mind that the storage volumes in this study were sized to minimize at the most the discharge from the system, while some user might have more discharge flexibility in practice. We also saw that the suggested total storage volume was oversized, particularly for the first 10 years or so. In practice, it would be possible to reduce the storage volume by sizing the equalizing tanks according to average net inflow and occasionally discharge a certain volume as needed (e.g. during extremely wet years). Such a sizing approach minimizing the total storage volume would still lead to a significant reduction in off-site treatment needs and could even be more attractive for industries with limited storage space. Alternatively, under climate with large interannual precipitation, ZLD beds sized to minimize discharge during wet years may result in the systems often lacking water during dryer spells. Plant species chosen for ZLD system should be able to sustain short periods of drought, but a reliable source of water for occasional irrigation of the planted bed may be necessary to maintain long-term plant growth and health.

Finally, biomass valorization is an important consideration: if willows or other woody species are to be used in a ZLD system, an option for valorization of the biomass produced (e.g. using fragmented stems as mulch in other locations, biofuel production) should be available. For example, shredded willow stems make excellent mulch, that has multiple uses (Lemieux et al., 2000), dried stems can be used as a material in different types of walls, barriers and screens (Lachapelle et al., 2019), and in some locations, the combustion of woody biomass can serve as an energy source (Langholtz et al., 2019). Although translocation of contaminants to aerial parts of the willows is expected to be low or null if water is pretreated, samples of stems could be analyzed prior to biomass valorization depending on the projected use.

4.4. Adaption to other climates and wastewaters

Willows are not adapted to every region of the world. They are naturally distributed mainly in the northern hemisphere (Argus, 1986) and are considered an invasive species in some southern locations like Australia, South Africa, and Argentina (Stokes, 2008; Henderson, 1991; Serra et al., 2013). A recent study concluded that *Salix humboldtiana* would be a good candidate for ZLD wetlands in Colombia (Moreno et al., 2019). It would be interesting to expand future research to alternative species with a high ET potential adapted to other climates, such as bamboo (*Bambusa* sp.) or giant reed (*Arundo donax*) keeping in mind that annual ET needs to exceed annual rainfall per surface area for the technology to be technically feasible. Again, a minimal ratio of 1.5 (willow bed ET versus rainfall, per m^2) seemed necessary in this study, although this value might vary in a different climate.

Regarding the applicability of the general technology to other geographic regions, climate is probably the factor that will dictate the end result of the design process. Our study has confirmed the feasibility of using a willow wetland to achieve ZLD in relatively unfavorable conditions (short growing season and high precipitations, occurring mainly during low ET period) which suggests that most climate could sustain this technology. As a general reference, we can presume that drier climate, or were annual temperature and/or rainfall variation low, and/or the vegetation growing period is longer, land area and storage volume needs would be significantly reduced because of greater overall ET (and therefore ET/rainfall ratio) and/or less variable hydraulic loading. However, other considerations could emerge in particularly arid climates, such as water scarcity, preference for technologies allowing water reuse and the lack of a constant and sufficient water supply for plant optimal growth.

The use of a willow bed ZLD treatment system, and therefore the design considerations pointed out in this study, is not limited leachate management. Other industrial wastewaters having sometimes low concentration of problematic contaminants (e.g. dyes in the textile industries or tannin and lignin in winery effluent; Vymazal, 2014) or stormwater and agricultural runoffs, that have highly variable volumes and represents a major concern in many parts of the world (Walsh et al., 2012; Daniels et al., 2018), represent other potential applications of the technology.

4.5. Study and technology limitations

The hydrological model presented here shows that, in the climate tested, the water storage volume required might be the most limiting aspect of this technology. Most of the annual rainfall occur during cold months were ET is low (typically autumn and spring) and therefore leachate most be stored to be used during the few summer months. Inversely, most of the ET occur during the three hottest summer months, and water demand is so high at this point that leachate accumulated in the storage unit during the cold months is not always sufficient to maintain a minimal water level in the willow bed.

Another aspect of any ZLD treatment system that was not tested in our model but that should be investigated to better assess the potential of the technology is the expected lifespan of the evapotranspiration wetland. Even if concentration of contaminants and/or salts in the leachate are low, an accumulation might occur after several years raising questions about the fate of any such those contaminants and their effect on plant health (Brix and Arias 2011). In the specific case of the reference site presented here, the treatment efficiency of the pre-treatment step was generally high (Levesque et al., 2017), and after seven years of operation, the trees in the willow bed showed no phytotoxic symptoms and no contaminant accumulation could be detected in the substrate (Frédette et al., 2019a). Furthermore, the cultivar used in this willow bed (*S. miyabeana* 'SX67') appears to be tolerant to the raw leachate produced on site (Frédette et al., 2019c). We could therefore estimate that the expected life span of the willow bed, in this case, exceeds 10 years. In a ZLD system with willow bed, the ET wetland is sized based on water volume, and not contaminant loadings, as is the case for a treatment wetland (Kadlec and Wallace, 2008). Therefore, the ET wetland will likely be well over-sized in terms of contaminant treatment capacity, and the life span of the wetland should then exceed, or at least equal, the typical life span of treatment wetlands (40–50 years; Kadlec and Wallace, 2008). Exceptionally, it is possible to pump out the contaminated or high-salinity water out of the willow bed to increase its life span. Ultimately, when a willow bed attains its maximal life span, the substrate should be removed and treated as recommended depending on the local regulations and contaminant concentration.

Another consideration, that is also a limitation of our study, is the weather (particularly rainfall) time frame used for sizing the compartments of the system. Using meteorological data from a shorter or longer period could have led to significantly different sizing. Furthermore, past rainfall data are not predictive of future conditions, considering the expected effects of climate change on local precipitation. For instance, for the region of the reference site of this study, annual rainfall increased by 130 mm (about 15% increase) from 1960 to 2013 (MDDELCC, 2017) and climate change scenarios predict a continuous increase in the future (Ouranos, 2015). One way of dealing with such factors would be to run simulations with predicted future data sets for longer periods (e.g. 50 years). However, this also enhances the need for flexibility in the design of a ZLD system where inflow comes from precipitation. Consequently, it is crucial to include a risk reduction strategy in the design, based on options such as irrigation or discharge, while reserving space for future expansion of the wetland or storage tanks.

5. Conclusion

Our results suggest that treating leachate with an ZLD system using an evapotranspiration willow bed is feasible, even in a climate with great seasonal temperature variations, a short growing season and relatively high annual precipitation (southeastern Canada). Although it requires a considerable land area and water storage volume, our results suggest that design and management decisions can significantly reduce those requirements. All those aspects (e.g. willow bed design, acceptable discharge frequency, collection tank volume, combined or separated storage tanks, etc.) need to be considered to ensure proper sizing of the system compartment and to increase the overall efficiency. We showed that an organic substrate with a high fertilization rate makes is possible to reduce the size of willow bed required. A 2-year coppicing cycle, meaning that every year, one half of the willows are cut back, should be used to maintain maximal ET rate. Simulating the operation of a system over a 20-year period highlighted the importance of designing a flexible system when treating leachate or other wastewater generated by rainfall. Modeling the temporal variation of both climate and ET is essential to achieve a flexible design. Although the operation of a willow bed is relatively simple compared to other ZLD technologies, water flux in the system must be monitored and managed to ensure maximal ET and minimize overflows. ZLD systems using a willow bed constitute a solution to some of the current limitations of typical ZLD treatment systems, such as high operational and energy costs and the difficulty of treating recalcitrant organic molecules, and our study constitutes a first step in extending this technology beyond domestic wastewater treatment. Future research on ZLD wetlands could focus on (1) quantifying the effect of fertilization on ET for several species used in ZLD wetlands, (2) determining the quantitative relationship between ET and substrate physical properties, (3) including a model of the fate of contaminants in the design system, (4) producing a life-cycle analysis, (5) investigating alternative species options and (6) testing the full-scale application of such a system based on the general design presented here.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

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