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A case study on the use of constructed wetlands for the treatment of wastewater as an alternative for petroleum industry

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RESUMEN

Los sistemas de humedales construidos están siendo ampliamente usados en el tratamiento de aguas residuales como herramienta en la defensa de la contaminación del agua y la necesidad de cuantificar el grado al cual la contaminación ha llegado a impactar el medio ambiente es un reto. Por lo tanto, es necesario desarrollar herramientas para determinar el impacto ambiental de contaminantes y monitorear la recuperación ecológica de un sitio luego de su remediación. Para evaluar la tecnología de los sistemas de humedales construidos, se implemento un proyecto piloto para el tratamiento de aguas residuales, usando diferentes sistemas de humedales construidos estableciendo ventajas y desventajas en cada caso, analizando su comportamiento y eficiencia en la renovación ambiental del agua, desarrollando un proceso de descontaminación usando plantas acuáticas y terrestres en los humedales experimentales, monitoreando el proceso de tratamiento, desarrollando, evaluando y mejorando los criterios para su diseño y operación y transferencia de tecnología. Los sistemas de humedales construidos representan una alternativa para la industria del petróleo, debido a que son igual o mas efectivos en la remoción de contaminantes que aquellos usados durante el tratamiento de otros tipos de aguas contaminadas, llegando a ser importantes para su manejo en una variedad de instalaciones, tales como refinería, pozos de gas y petróleo y estaciones de suministro, donde Los sistemas de humedales construidos suministran beneficios en la predicción de la calidad del agua cuando son diseñados y mantenidos apropiadamente.

PALABRAS CLAVES

sistemas de humedales construidos ; aguas residuales; tratamiento; ambiental; petróleo; tecnología

ABSTRACT

Constructed wetlands are increasingly being employed for wastewater treatment, which is the last line of defence against water pollution, and the need to quantify the degree to which the pollution has actually impacted the environment has become paramount. Therefore, there is a need to develop tools for determining the environmental impact of pollutants and monitoring the ecological recovery of a site after remediation. To evaluate the constructed wetland technology, a municipal wastewater demonstration project was implemented, using different types of constructed wetland systems, for the wastewater treatment to evaluate relative advantages and disadvantages of these types of systems, analyze their behavior and efficiency in water environmental renovation, develop a wastewater treatment using vegetation species in the experimental wetlands, monitor

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the treatment process, develop, evaluate and improve basic design and operation criteria, and transfer technology. Constructed wetlands represent an alternative for the petroleum industry, because they are equally or more effective at removing pollutants than those used during the treatment of other types of wastewater, becoming important to manage process wastewater at a variety of installations, such as refineries, oil and gas well, and pumping stations, where constructed wetlands provide predictable water quality benefits when properly designed and maintained.

KEYWORDS

Constructed wetlands; wastewater; treatment; environmental; petroleum; technology

INTRODUCTION

Treatment wetlands have been recognized as providing many benefits including water supply and control, mining, use of plants, wildlife, fish and invertebrates, integrated systems and aquaculture, erosion control, gene pools and diversity, energy, education and training, recreation and reclamation (e.g., Peterson, 1998; Drizo et al., 1997; Richardson et al., 1997; Vymazal et al., 1995; Crites, 1994; Brix, 1987; 1994). Constructed wetlands (CWs) systems are becoming increasingly important as an effective and low-cost in construction and maintenance alternative for wastewater treatment around the world since the early 1970s. Over the past 15 years, the petroleum industry has been one of the pioneers using CWs to treat a variety of wastewater at a variety of installations, including refineries and fuel storage tanks, where CWs provide predictable water quality benefits when properly designed and maintained. However, there is scarce information (e.g., Ji et al., 2002) about the purification of CWs for the main pollutants of petroleum industry effluents.

CWs can be defined as transitional environments between terrestrial and aquatic systems with the main purpose of contaminant removal from wastewater systems. They are usually categorized by origin, hydrologic type and vegetation type. According to the origin they can be natural (a pre-existing wetland that is incorporated into the treatment system), constructed (a completely artificial wetland built specifically for wastewater treatment) or hybrid (has both natural and CWs as part of the treatment system). The hydrologic type can be either free water surface FWS or subsurface flow SF (the term subsurface flow is a generic term that includes all types of systems where the wastewater is below the ground level). The vegetation type includes marsh forest, floating aquatic plants, as well as other less common categories, and plants of several categories can be present, especially in natural treatment wetlands.

The major components of CWs are vegetation, hydrology, substrate and microbial populations. Different studies (e.g., Breen & Chick, 1995; Abe et al., 1993; Ozaki & Abe, 1993; Brix, 1987) evaluate and compare the effectiveness of vegetation species in pollutant removal from wastewater. The vegetation of CWs plays a very important role during the treatment process, because it helps in supplying oxygen to the microorganisms in the rhizosphere, increase water loss by evapotranspiration, reduce the amount of nutrients in the system by uptake, and perhaps more surface area in the rhizosphere for the miscroorganisms (Brix, 1987). The performance of wetland systems to remove pollutants from wastewater can be improved by using suitable substrates (e.g., Sakadevan & Bavor, 1998; Drizo et al., 1997; Johansson, 1997; Wakatsuki et al., 1993; Ciambelli et al. 1985; Blanchard et al., 1984).
This paper looks at different aspects of CWs with the aim of promoting how such systems can be integrated into wastewater treatment. The main purpose of this research was to develop a

wastewater treatment technique in combination with recycling and amenity functions in order to evaluate and compare the effectiveness of bed filter materials along with terrestrial and aquatic plants in the water environmental renovation of municipal stream at the Kawaguchi Park, which is located in Matsue City, Shimane Prefecture (Japan) (Figure 1). However, this study is also concerned with the importance to consider the constructed wetland systems as a low-cost of construction and maintenance and energy-saving alternative for reducing the pollutants of primary importance to the petroleum industry.

CONSTRUCTED WETLANDS

CWs are man-made systems that provide a low-cost natural treatment for wastewater from towns and small cities, urban storm water runoff, livestock producers, failed septic tank drain fields, mine drainage, landfills, and many types of industry, accomplishing water quality improvement through a variety of physical, chemical and biological processes operating independently in some circumstances and interacting in others. CWs remove high levels of particulates, as well as some dissolved contaminants. Therefore, it is likely to have a significant impact on sediment, nutrients, heavy metals, toxic materials, floatable materials, and oxygen demanding substances and oil and grease. Untreated petroleum industry wastewater contain many of the same pollutants as municipal wastewater such as the example described in this study, as well as oil and grease, different hydrocarbons, phenolics, sulfides and metals. Knight et al. (1999) briefly describe the features of inquiries of petroleum companies that developed full and pilot-scale constructed wetland treatment projects to provide water quality improvement.

Categories of wetland treatment systems

CWs are classified according to their mode of operation as surface-flow, horizontal flow, and vertical down flow or vertical up flow type. On the other hand, CWs for wastewater treatment May be classified according to the life form of dominating macrophyte: free-floating macrophyte-based systems, submerged macrophyte-based systems, and emergent macrophyte-based systems. Wetland treatment systems can be categorized as follows according to the flow pattern (Figure 2).

Systems with free-water surface (FWS). FWS systems (Figure 2a) consist on a channel surrounded by a barrier of pounded wastewater and substrate to support the growth of rooted emergent vegetation. The shallow water depth, low flow velocity, and presence of the plant stalks and litter regulate water flow and, especially in long, narrow channels, ensure plug-flow conditions (Reed et al., 1995). Emergent vegetation grows and wastewater is treated as it flows through the vegetation and plant litter. It includes an open-water surface as part of the system. The advantage of FWS systems is the low risk of clogging, but treatment efficiency for some components is sacrificed because of lower substrate to pollutant interactions. The open-water wetland has a small substrate to root the plants while the hydroponics wetland does not. Hydroponics wetlands use floating plants, and the root system alone as a filter, depth of water bed is very shallow (50mm), and particulate nutrient are trapped by the root system and decomposed.

Systems with horizontal subsurface flow (HSF). HSF systems (Figure 2b) are called horizontal flow, because the wastewater is fed at the inlet and flows slowly through a porous medium under the surface of the bed in a more or less horizontal path until it is collected before leaving via level control arrangement at the outlet. They consist on an excavated and usually lined shallow basin, containing a substrate to a depth of approximately 0.6m, and emergent aquatic plants. The wastewater will come into contact with a network of aerobic (around roots and rhizomes, leaking oxygen into the substrate), anoxic and anaerobic zones, and during its passage through the rhizosphere, it is cleaned by microbial degradation and physical/chemical processes (e.g., Brix, 1987). The water level is maintained below the surface of the substrate, which minimizes the risk of exposure to people and animals and greatly reduces mosquito breeding. HSF systems are the most common CWs used for small flows and are often used for individual homes, small clusters

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of houses, or resorts.

Water quality improvement performance in treatment wetlands

In CWs, inflow water containing particulate and dissolved pollutants slows and spreads through a small area of shallow water and emergent vegetation. The particulates are typically measured as total suspended solids (TSS), which tend to settle and are trapped in a substrate due to lowered flow velocities and sheltering from wind, and contain biochemical and chemical oxygen demanding (BOD and COD) components, hydrocarbons and other organics, trace metals, and fixed forms of total nitrogen (TN) and total phosphorus (TP).

CWs have a wide variety of engineering designs, wetted areas, flow rates, inflow water qualities, plant communities, hydrologic regimes, effluent limitations, and monitoring requirements, which have been summarized by Knight et al. (1999). The advancement of CWs' technology and the ability of designers to take advantage of wetland processes in predictable treatment systems hinges on the ability to summarize treatment wetland data sets into a small number of defining relationships, which include loading rates and removal efficiencies, regression equations, and first-order mass balance equations. Currently, models represent different levels of complexity can generally be calibrated with treatment wetland data, providing a reasonable approximation of performance for a wide range of pollutants in CWs, although it does not account for adaptation trends, the effects of dissolved oxygen and pH on performance, and many other factors that affect the destination of pollutants in CWs. Additional advances in providing more complete descriptions of CWs' behaviour are dependent upon analysis of data from comprehensive research projects, including pilot studies. Different operative wetland processes contribute the removal, conversion or storage of hydrocarbons and other chemicals in treatment wetlands, including volatilization, partitioning to sediments, biofilms and humics, mass transfer to sorption/degradation sites, biodegradation, photodegradation, and plant and animal uptake (Knight et al., 1999).

MATERIALS AND METHODS

Design of constructed wetlands

A general guidance for planning of CWs was followed in this study to identify their potential sites, estimate how to connect them to the area tributary, select types of CWs to be used, determine their required volume, integrate them into site arrangement, determine their inlet and outlet features, and carry out their maintenance.

CWs for experimental demonstration were located at the Kawaguchi Park (Figure 3a), consisting of channels with vertical walls, each 10,0 m long, 0.4 m wide and 0,3 m depth, which provide 4,0 m^2 of surface area (Figure 3b). They were filled with alluvial rhyolitic gravel (channel-1), mordenite (channel-2), a mix of charcoal, soil and mordenite (channel-3), and water (channel-4), and planted with rooted emergent terrestrial and/or aquatic macrophyte species. After plantation, the municipal stream was pumped into an inlet tank (0.8m^2) and distributed to the channels, which were initially filled by one-month period for plant acclimatization. The flow volume was adjusted to 5.6 lt/min with 2 m^3/m^2/day as water loading. The detailed description and anatomy of the CWs used in this study are listed in Table 1 and shown in Figure 4, respectively, and their distribution and details of the vegetation species is illustrated in Figure 5.

Design of plant bed filters

Plant-bed filters for the cultivation of terrestrial and/or aquatic plants were used for the experiment, consisting on substrates of gravel, mordenite, and a mix of charcoal, soil and mordenite. Different CWs such as FWS or HSF systems and biogeofilter (BGF) were considered. Three types of CWs were used in all channels, except in channel-4 (a hydroponics biofilter (HBF) after Aizaki & Nakazato (1997)). The biogeofilter (BGF) method consists on CWs, which have an excavated and usually lined shallow channel, containing substrate media to a depth approximately 26cm, and emerged terrestrial and/ or aquatic plants.

Wetland vegetation

The vegetation species tested in the experiment from May to November of 1999 and from April to June of 2000 to evaluate their efficiency in removing N and P from the wastewater are listed in Table 1. In the first phase of the experiment, each channel was planted with 8 species of terrestrial plants, 1 species of aquatic/terrestrial plant, and 1 species of aquatic plant, being difficult to find the appropriate conditions for the good growth of the plants. In the second phase of the experiment, 11 species of terrestrial plants were sowed, which evidenced a better development in each one of the channels, although similar results as in the previous stage were obtained. The selection of plant species for the CWs took into consideration the water fluctuation likely to occur in the wetland. Plant acclimatization lasted one month after seeding.

Sampling and chemical tests

Grab samples were taken from the influent and effluent channels from summer to autumn 1999 and from spring to summer 2000 and introduced into a 1 lt polyethylene bottle and packed in crushed ice between sampling and analysis to avoid biodegradation or volatilization. When the analysis was no immediately done, samples were kept at 4ºC. DO, pH, conductivity, turbidity, temperature and salinity were measured in the field. Wastewater additions and treatment performance from supplied urban channel was collected in an inlet channel. Samples from wastewater inflow and channels were taken to measure wastewater performance. Sampling was performed once a week and analyzed with standard methods (APHA, AWWA and WPFC, 1989) during 5 days in order to estimate BOD_5, SS, COD, TN, DTN, TP, DTP, PO_4-P, NH_4-N, NO_2-N and NO_3-N. 20 minutes or 1 hour of hydraulic retention time (HRT) for every channel. TP and DTP were determined after alkaline peroxydisulfate digestion in an autoclave at 120ºC for 45 min by the method of Menzel & Cowin (1965); PO_4-P after the ascorbic acid method (Murphy & Riley, 1962), NH_4-N after the method of Otsuki & Sekiguchi (1983), NO_3-N by colorimetry after the cadmium reduction method of Strickland & Parsons (1962), NO_2-N by colorimetry after sulphanilamid and N-(1-naphthyl) ethylendiamine, BOD after 5-day BOD_5, COD after titrimetric method with K_2MnO_4 and SS after GF filter gravimetric. Analytical methods are shown in Table 2. The chemical analyses phase was carried out from April 14 to June 26 of 2000.

RESULTS AND DISCUSSION

The results obtained were within the standard limits in TN, TP, SS and BOD_5. For COD the results were not very good because the procedure used in the analysis was not the correct.

Physical conditions in the field

The physical conditions measured in inflow water and outflow water of each channel are shown in Table 3. Water temperature ranged in each month. High values of 25-30ºC were observed in August and September of 1999. All experiments were carried out above 10ºC. pH values ranged from 6.4 to 7.3 in inflow water and from 6.8 to 8.6 in outflow of each channel, and increased by pass through the channels. DO concentrations ranged from 7.9 to 11.1 mg/L in inflow water and from 7.7 to 12.8 mg/L in outflow water of each channel, and increased by pass through the channels. Conductivity showed high values of 15.3-24.4 and 11.6-20.9 mS/cm in August and October, respectively, in inflow water. Salinity showed a same tendency. These high conductivity and salinity affected the growth of plants. Table 4 summarizes the results of mean concentrations and removal percentage of NH_4-N, NO_3-N, NO_2-N, TN, TP, PO_4-P, TBOD and SS, between 1999 and 2000. Figure 6 shows percentages of removal of NH_4-N, TN, TP, PO_4-P, TBOD and SS, and concentrations of NO_3-N and NO_2-N, respect to the date, between 1999 and 2000.

Removal of Nitrogen (see Table 4 and Figure 6)

Wastewater generated from municipal, agricultural and industrial sites creates NH_4-N that subsequently mixes into lakes, rivers and drinking water reservoirs, decreasing the DO required for the aquatic life and accelerating the corrosion of metals and construction materials. Nearly all treatment wetland studies have reported

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reductions in TN and N. Petroleum industry data for N are summarized in Knight et al. (1999). N removal rate constant values for these wetlands are comparable to or higher than values for other treatment wetlands. Treatment wetland removal of all major N forms is sensitive to temperature.

Ammonium (NH_4-N). In 1999, the NH_4-N removal trends tend to increase from August to October, with a small decrease from August to November, except for treatments in channel-2 and channel-4 that show a reversal behaviour from September to November. In 2000, the removal trends of NH_4-N increase from April to May, decreasing from May to June, except for treatment in channel-1, which shows a reversal behaviour. The mordenite substrate tends to be the best for NH_4-N removal with the highest removal percentages of 87% (Nov/99) and 94% (May/00), followed by gravel, with 69% (Oct/99) and 84% (Apr/00), and the mix of charcoal, soil and mordenite, with 63% (Oct/99) and 85% (May/00). Channel-4 shows the lowest NH_4-N removal, with 53% (Nov/99) and 42% (May/00).

Nitrate (NO_3-N). In 1999, the NO_3-N concentrations show similar trends for all treatments, decreasing from August to November, except for treatment in channel-3, which shows the lowest concentration of NO_3-N (3 mg/L) in October. In 2000, these concentrations decrease from April to May, increasing from May to June, except for channel-3 treatment that shows a decrease from April to June. The highest concentrations were reached in April, with 1216 and 622 mg/L of NO_3-N, for channel-2 and channel-3 treatments, respectively. Treatments in channel-1 and channel-4 show the lowest concentrations, reaching in May 66 and 212 mg/L of NO_3-N, respectively.

Nitrite (NO_2-N). In 1999, the NO_2-N concentrations show similar trends for all treatments, decreasing from August to October, with a small increase from October to November, except for treatment in channel-4, which shows an increase from August to September, decreasing from September to November, and reaching the highest concentration of NO_2-N (195 mg/L) in September. In 2000, these concentrations decrease from April to May and increase from May to June. The highest concentrations of NO_3-N were reached in April, with 1507 mg/L in channel-1, followed by channel-2 (1195 mg/L), channel-3 (697 mg/L), and channel-4 (492 mg/L), respectively.

Total Nitrogen (TN). In 1999, the TN removal trends tend to increase from August to October, decreasing from August to November, except for treatments in channel-2 and channel-4, which show a reversal behavior from September to November. In 2000, the efficiency in TN removal increase from April to May, decreasing from May to June for treatments in channel-1 and channel-2, and increasing from April to June for treatments in channel-3 and channel-4. In the first phase of the experiment, channel-3 treatment showed the best efficiency in TN removal with the highest removal percentages of 54% (Oct/99), followed by channel-1, 48% (Oct/99), channel-2, 39% (Sep/99), and channel-4, 30% (Sept/99). In the second phase of the experiment, channel-3 and channel-4 treatments showed the best efficiency in TN removal, although the treatments in channel-2 and channel-1 showing the highest removal percentages of 63% (May/00) and 53% (May/00), respectively.

Phosphorus Removal

(see Table 4 and Figure 6)

CWs are capable of absorbing new P loading and could provide a low cost alternative to chemical and biological treatment. P interacts strongly with CWs' soils and biota, which provide both short-term and sustainable long-term storage of it. TP performance data from petroleum industry treatment wetlands are summarized in Knight et al. (1999). Reductions in TP are significant. Loadings are high compared to other treatment wetlands.

Total Phosphorus (TP). In 1999, the TP removal trends show a small decrease from August to October, abruptly decreasing from October to November, except for treatment in channel-1, which shows a reversal behavior from August to October. In 2000, the treatments in channel-2 and channel-4 show TP removal trends, decreasing from April to May and increasing from May to June. A similar behavior is shown by the treatment in channel-1, although it abruptly decrease from April to May. The TP removal trend in channel-3 shows a reversal behavior respect to that in channel-2 and channel-4. A mix of charcoal, soil and mordenite substrate tends to show the best efficiency in TP removal with the highest removal percentage of 71% (May/00), followed by gravel, 56% (Jun/00), mordenite, 41% (Jun/00) and hydroponics biofilter, 26% (Jun/00). The lowest TP removal, with -240% (May/00), was obtained in channel-1, followed by channel-4, -204% (Nov/99), channel-2, -194% (Nov/99), and channel-3, -99% (Nov/99).

Phosphates (PO_4-P). The best performance for removal of PO_4-P was observed using a mix of charcoal, soil and mordenite as a substrate, followed by that showed by gravel and hydroponics biofilter treatments, respectively, with a lowest efficiency for mordenite treatment. In 1999, the PO_4-P removal trends show an increase from August to September, with a decrease September to November, for treatment in channel-1 and channel-3, although the first one abruptly decreases from October to November. The treatment in channel-2 shows an irregular trend, with a decrease form August to September, increasing from September to October, and abruptly decreasing from October to November, reaching the lowest PO_4-P removal percentage of -394% (Nov/99), followed by channel-1, -280% (Nov/99). Channel-4 shows a trend that decrease from August to October, with a small increase from October to November. In 2000, channel-1 and channel-4 show trends that decrease from April to May, increasing from May to June, reaching the lowest PO_4-P removal percentage of -134% (channel-4), while treatment in channel-2 shows a reversal behavior. The treatment in channel-3 shows a trend that increase from April to June.

Organic Matter Removal (see Table 4 and Figure 6)

Total Biochemical Oxygen Demand (TBOD). The generation and return of five-day BOD (BOD_5) results from the death of wetland macrophytes and microorganisms attached to the submerged solids. These combined processes will produce elevated BOD_5 concentrations in CWs that typically decrease along the wetland flow path from inlet to outlet, down to the background level. Temperature apparently plays a minor role in the net removal of BOD_5 in treatment wetlands. No published petroleum wastewater data set are currently available to fully calibrate models for BOD_5 reduction.

In this study, in 1999, the TBOD removal increase from August to October, decreasing from October to November, except for treatment in channel-2, which show a decrease form August to September, increasing from September to November. In 2000, treatment in channel-2 and channel-3 show TBOD removal trends that increase from April to May, decreasing from May to June, while treatments in channel-1 and channel-4 display a reversal behavior. In the first phase of the experiment, the best efficiency in TBOD removal tends to be shown by channel-3, with the highest removal percentage of 73% (Sep/99), followed by channel-2, 51% (Nov/99), channel-1, 48% (Sep/99), and channel-4, 41% (Sep/99). In the second phase of the experiment, channel-2 showed the best performance, reaching the highest TBOD removal, with 63% (May/00), followed by channel-3, 49% (May/00). Channel-4 showed the lowest TBOD removal, with -16% (May/00).

Suspended Solids (SS). Treatment wetlands are typically efficient in reduction of SS concentrations, along the flow path from inlet to outlet, down to the background level. Temperature apparently plays a minor role in SS reduction. The removal of SS has not been the main focus of petroleum industry treatment wetland projects.

In this study, in 1999, the SS removal shows increasing trends from August to November for each treatment, except for that in channel-3, which decreases from October to November. In 2000, for channel-1 and channel-2 it shows a decrease from April to June, while for channel-3 and channel 4 it shows a decrease from April to may, increasing from May to June. The best

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performance in SS removal was shown by channel-1, with the highest removal percentages of 87% (Nov/99), followed by that in channel-2, 89% (Nov/99), channel-4, 84% (Nov/99), and channel-3, 77% (April/00). Channel-4 showed the lowest SS removal, with -135% (May/00).

Effects of structure of substrate

CWs are capable of significant metals removal, and different physical-chemical properties of substrates affect metal mobilization-immobilization processes. According to Grambell (1994), important substrate physical properties include particle size distribution and, to some extent, the type of clay minerals present, and substrate chemical properties affecting these processes include redox potential, pH, organic matter content, salinity, and the presence of inorganic components.

As mentioned before, we used different substrates in the experiment: gravel (channel-1), mordenite (channel-2), a mix of charcoal, soil and mordenite (channel-3), and water (channel-4). The substrate let the water flowed in a "S" trajectory, which allowed the oxygenation of the water and at the same time it entered to the substrate, the level of the substrate with relationship to the water was adapted with plants root that could touch the water. The behavior of mordenite (zeolite group) substrate was good compared with the others in relation with N removal. Mordenite can be mixed with other materials, which allow hydraulic conductivity to vary significantly, and, therefore, its hydraulic conductivity permits use it in a wide range of substrates. Zeolite's high cation exchange capacity allows for nutrients such as ammonium, required for microbial metabolism, to be attached onto the structure of the zeolite. The presence of extra nutrients increased the biodegradation of contaminants entrapped by the zeolite. This study showed that zeolites possess unique properties, such as structure and surface charge, which permit them contribute to the treatment of municipal wastewater.

Flower plants in wastewater treatment

Metals removal from wastewater by plant roots has been demonstrated in a number of studies. In particular, iron root plaque formation and emergent plant roots are important factors in biogeochemical processes in wetlands because iron, with organic compounds, composes the most important redox buffer system in wetlands, and because emergent plant roots contribute to the aeration of sediment, adsorption of heavy metals, oxidation of methane, and the diversity of microorganisms in the rhizosphere (Wang & Peverly, 1996).

In general, in this study, the N and organic matter removal through of the flower plants was very poor and for the P removal was null. Previous studies show that the efficiency in removal of BOD, TN and TP generally is independent of the vegetation species used during the treatment, but is sure that the plants provide the better conditions for sedimentation, diminish the risk of erosion, etc.

In the experiment, channel-1, channel-2 and channel-3 were sowed different species of plants, mainly with flowers, trying to find the appropriate conditions for their development and growth and also to find the species that adapted better to the system. The water was collected in an entrance channel and in this part of the channel it travelled below the substrate about 5cm from the surface, which was enough so that the plants reached to be in contact with the water and the other part travelled to the surface of the substrate reaching the oxygenation and entering inside the substrate again. In a first phase of the experiment, 13 species were sowed, although they didn't resist those conditions and, in a second phase of the experiment, aquatic plants such as Seri and Kreson were sowed and the results were good. Finally, 13 species of terrestrial plants were sowed and good results were observed for their development and growth. The plugging affects the continuous journey of the water, producing an accumulation of salt in the substrate that affect the growth and development of the plants.

Effect of water loading

Nitrogen, ammonium and phosphorus. During the experiment, the N and ammonium removal was not very high since it was affected by the loading rate, and when it was high the removal levels diminished. The hydraulic loading rate of the system varied with the season, generally being highest during spring where the system received a fair amount of rainwater in addition to the wastewater, and, therefore, high hydraulic loading rates are usually coupled with rather dilute wastewater and vice versa.

As the P removal in CWs happens always through of adsorption, plant absorption, precipitation and retention, it is probably that the major P removal was due to retention (by the substrate) and precipitation. In many cases the levels of phosphates in outlet was very high probably related to their accumulation in the substrate in several opportunities, covering the entrance channel, as well as the accumulation of algae or another class of materials that impeded the continuous entrance of water.

Organic Matter. The removal of BOD and SS was also affected by the variations in the levels of water loading, although the last one diminished notably when these variations were very high. COD represents the class of organic compounds that are susceptible to oxidation by a strong chemical oxidant under acidic conditions, and is numerically higher that BOD_5, because more organic compounds can be chemically oxidized that are degraded biologically. Limited information on petroleum wastewater treatment CWs indicates that COD is reduced at rates comparable to CWs treating other types of wastewater. Temperature effects on COD are typically minimal, although data are scarce. Trace organic removal from petroleum wastewater are problematic for treatment wetlands because organic compounds present at high concentrations may be potentially toxic to plants and microorganisms, and because different organic compounds found in the wastewater have different susceptibilities to aerobic and anaerobic degradation processes. However, most hydrocarbons are natural products and are biodegradable, and many of them are not toxic to organisms except at high concentrations and some are used as growth enhancers at low concentrations. Through natural processes, CWs produce a wide range of organic compounds, which may form complex molecules with metals and serve as an important mechanism to buffer redox reactions in CWs (Wang & Peverly, 1996). Knight et al. (1999) summarize data for organic compounds removal data from petroleum industry treatment wetlands, including refinery effluents, spills and washings, oil sand processing water, and production of water from natural gas processing.

Metal removal. Metal removal efficiencies of treatment wetlands are highly correlated with influent concentrations and mass loading rates (e.g., Stark et al., 1995). Treatment wetlands reduce acute and chronic toxicity to both cladocerans and fathead minnows in almost every case studies (American Petroleum Institute, 1998). The magnitude of toxicity reduction is typically inversely related to the wastewater loading rate and directly related to the effectiveness of mixing (water flow distribution) within the treatment wetland, and these general observations suggest that toxicity reductions in treatment wetlands are likely a secondary benefit of the myriad of pollutant removal processes in these complex biological systems (Knight et al., 1999). Additional research is required on the specific mechanisms of toxicity reduction in treatment wetlands.

Problems. There were several problems, because the channels were blocked with some been accustomed to suspend that they came from the main channel, but they were only corked one day. A worm also invaded channel-4 (Seri) and all plants died. Another obstacle occurred when levels of salinity increased affecting the growth of the plants, which didn't show a good development. Some inconveniences also presented in channel-1 because the entrance channel was constantly blocked with some residuals and algae, which impeded the continuous entrance of water affecting the treatment process.

CONCLUSIONS

CWs proved their high efficiency in removal of organic matter (BOD) and nutrients (N, P and

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SS), which was influenced by the loading rate (high levels of water loading affected the process notably). The removal percentage of TN, TP, ammonium, BOD and SS increased when plants combined with forms of animal life such as the bivalve *Curbicula japonica*, which is a very good wastewater cleaner that gave origin to an excellent ecosystem for improving the quality of water (e.g, Aizaki et al., 1998; Nakamura et al., 1988), which is very important taking into account that the municipal stream flowing through the experimental site is strongly influenced by the Ohashi River, which crosses the Matsue City shares a long fertile basin with Shinji Lake to the west and Nakaumi Lagoon to the east. The Shinji Lake is a mesohaline backish lagoon, and its freshwater derives mainly from the Hii River. Polyhaline water of the Nakaumi Lagoon, which opens on the Japan Sea, occasionally flows back into the Shinji Lake through the Ohashi River at its east end. *Curbicula japonica* inhabits the coast densely at depths < 4m in the Shinji Lake (Nakamura et al., 1988).

The continuous inflow of water allowed the oxygenation of the CWs, which favoured the development of a combination of physical-chemical and biological processes including sedimentation, precipitation, absorption of soil particles, assimilation for the plant tissue and microbial transformations. The temperature was inside the appropriate ranges so that nitrification and denitrification were carried out. The high levels of salinity affected the development of the plants, sometimes exceeding the limits, by which plants died.

According to the last years' tendencies, we decided to construct SSF-type wetlands rather than FWF-type, because such systems are believed to be more effective for treating wastewater. The experiment was carried out using Biogeofilter (BGF) and Hydroponic Biofilter (HBF) and different species of plants. BGF model showed the best environmental conditions, and the plants had an appropriate development and excellent adaptation to the substrate. Despite current usage patterns, tropical and subtropical climates hold the greatest potential for the use of CWs, whereas cold climates brought problems of icing and thaw.

The properties and capabilities of mordenite leave no doubt about why industrial and scientific communities are lured by zeolites, which tend to be more effective as substrate for the P and N removal from domestic wastewater than the other used materials. Testing also shown that nutrient enriched mordenite provides the proper environment for the biodegradation of entrapped organic contaminants. Mordenite hydraulic conductivity makes it ideally suited to serve as a permeable barrier. The variety in structures permits chemical selectivity, modification, and enhancement.

Use of CWs in developing countries such as Colombia could provide real economic benefits by providing biomass and supporting aquaculture, but in developed countries as Japan, the wetlands systems could become a habitat for wildlife and act as a tourist attraction for the community.

The benefits of CWs applied to the petroleum industry will include improvement of water quality and landscape amenity, recreational opportunities, creation of fauna and flora habitat, opportunities for storm water harvesting, sometimes combined with aquifer storage and recovery system, a degree of flood retention function, and community education designed to promote increased knowledge, improved skills and more positive attitudes about the environment. CWs can be developed into healthy ecosystems that are productive diverse and resilient, with a significant biomass of plants and microorganisms, which take up nutrients and provide habitat and food for animals, as well as different animal, plant and microbial species that promote materials transfer and decomposition of organic material. Treatment wetlands will be productive, diverse and resilient so they can continue to perform their pollutant removal function effectively, and healthy ecosystems, once established in CWs, will maintain themselves indefinitely through growth and reproduction of all species of plants and animals with minimal on-going maintenance.

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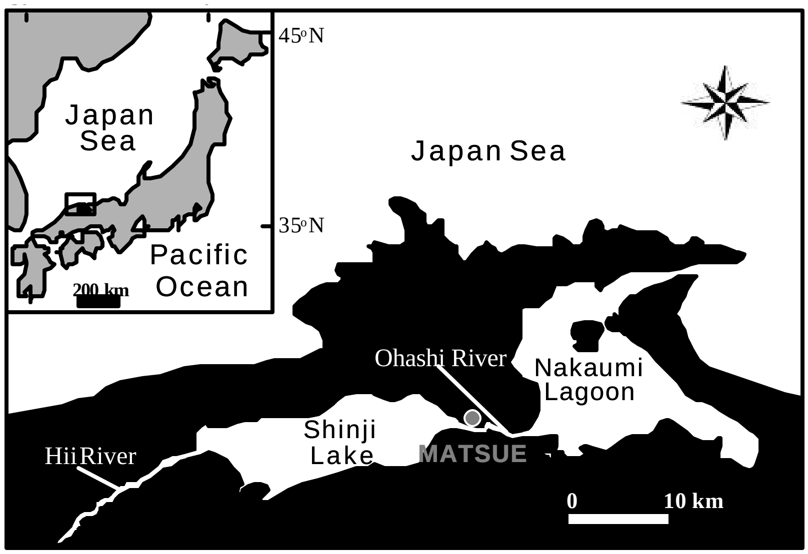

Figure 1. Map of geographic location of the Matsue City, on the west coast of Japan, which is crossed by the Ohashi River that connects the Shinji Lake and the Nakaumi Lagoon

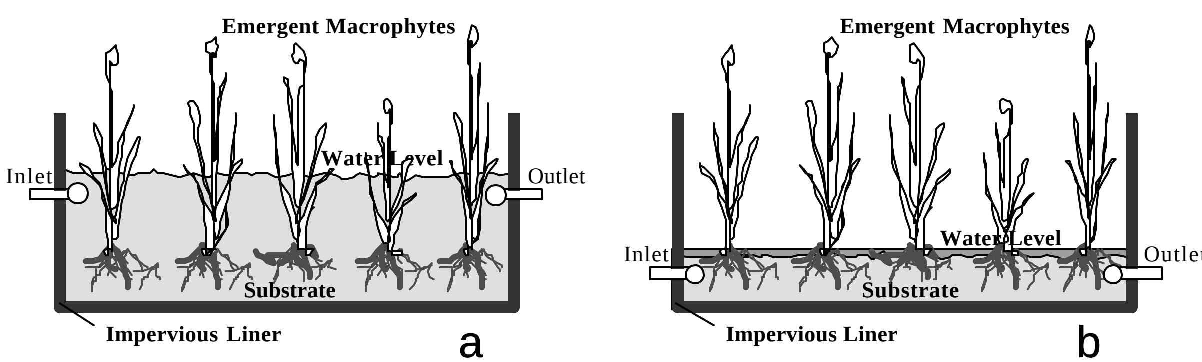

Figure 2. (a) Free-water surface system (FWS), where the water flows through leaves and stems. (b) Horizontal subsurface flow system (HSF), where the water flows through substrate and roots.

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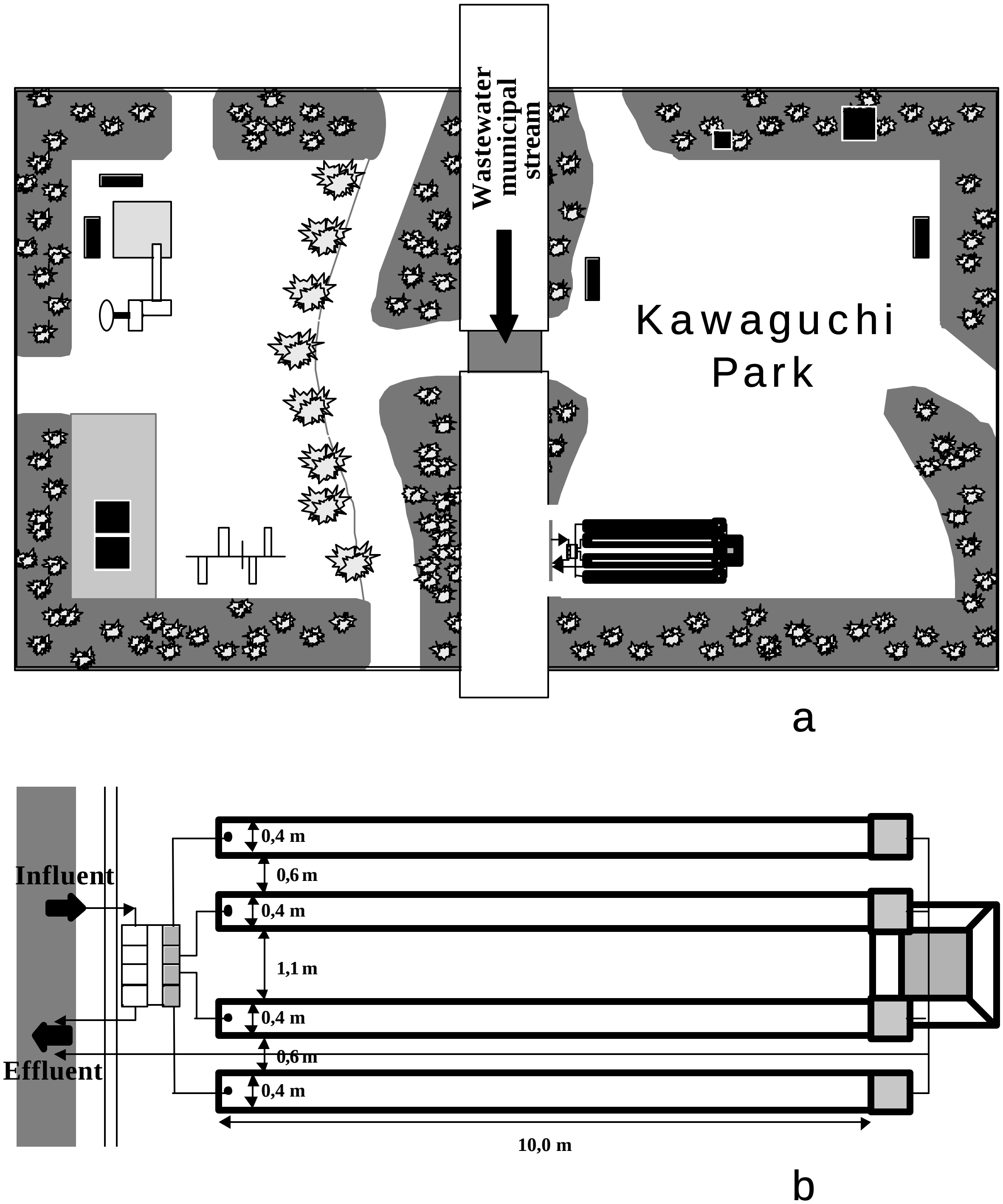

Figure 3. (a) Location map of the experimental site at the Kawaguchi Park, Matsue City, Shimane Prefecture (Japan). (b) General layout of the constructed wetlands used in this study, showing the distribution of channels (1, 2, 3 and 4).

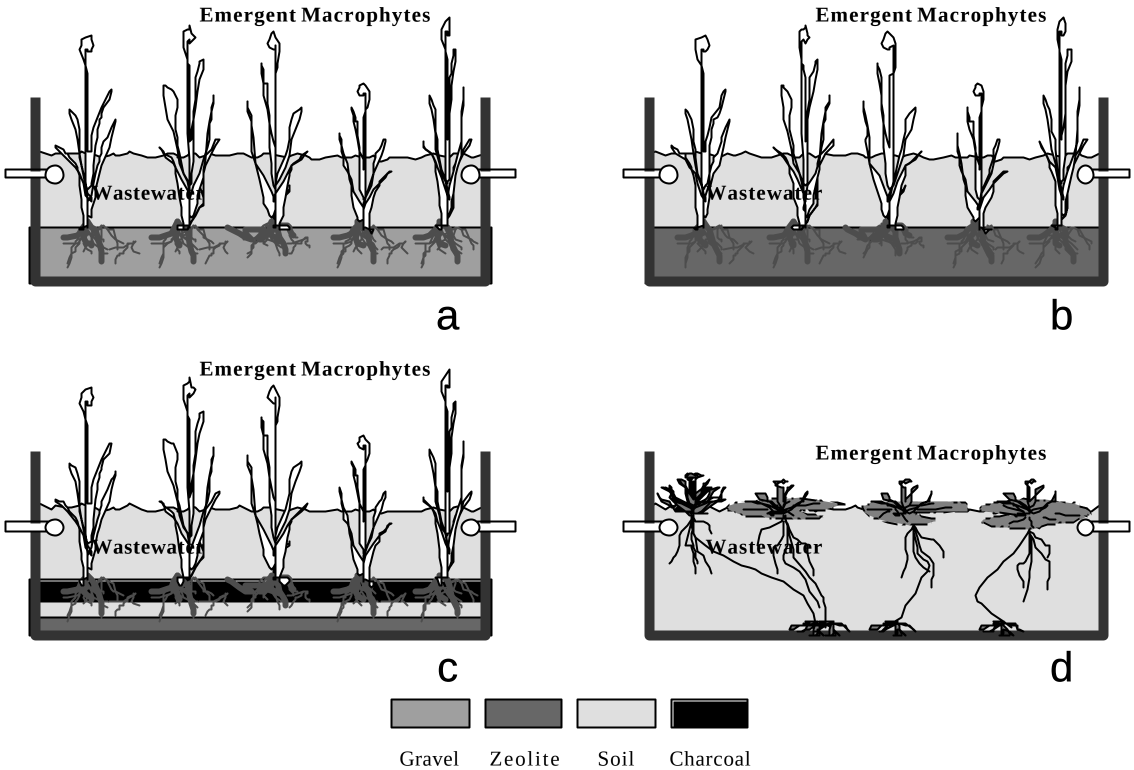

Figure 4. Constructed wetlands systems used in this study: (a) with gravel as substrate; (b) with mordenite as substrate; (c) with a mix of charcoal, soil and mordenite as substrate; (d) with no substrate (Hidroponic Biofilter).

Figure 5. Distributions of the channels (1, 2, 3 and 4) located at the Kawagushi Park, and details of the vegetation species (Impatiens sultanii and Seri) used for the constructed wetland systems of this study.

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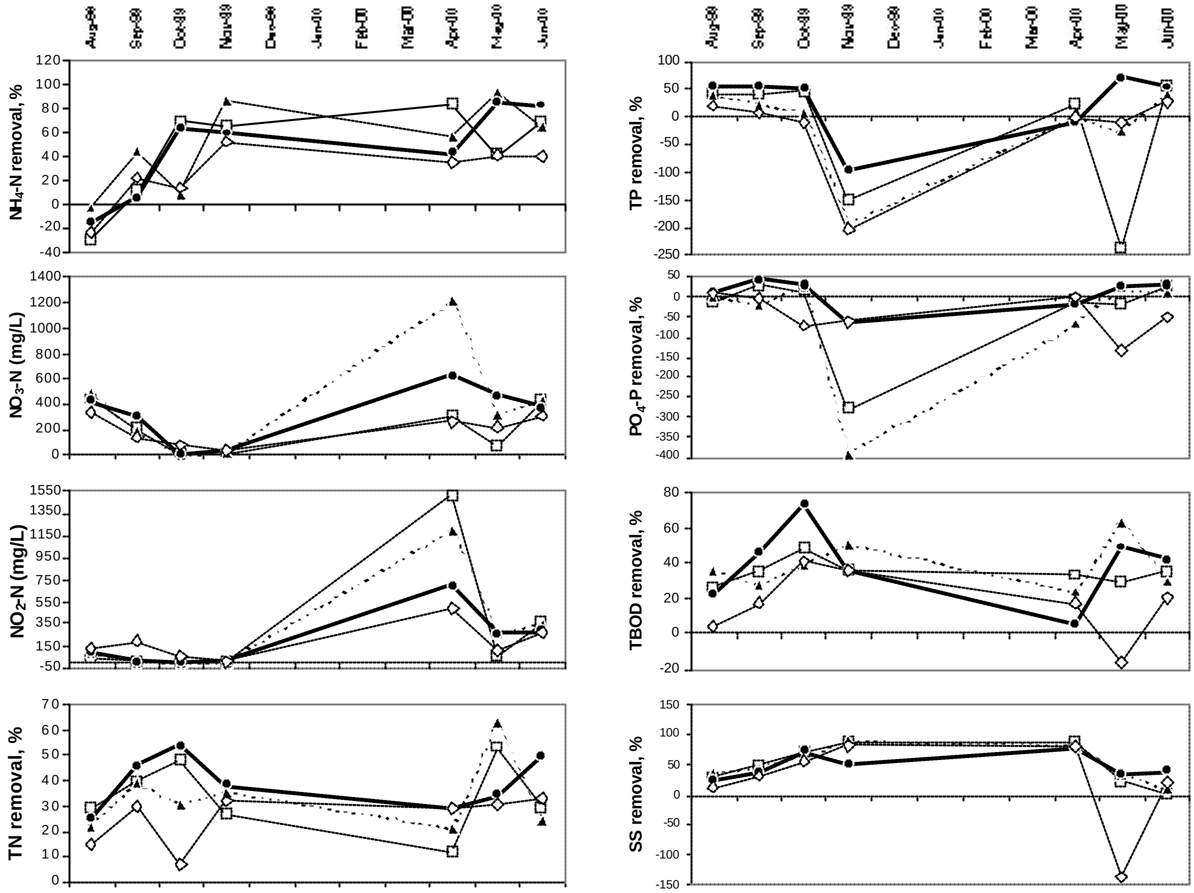

Figure 6. NH_4-N, NO_3-N, NO_2-N, TN, TP, PO_4-P, TBOD, and SS. Substrate: gravel (Channel-1); mordenite (Channel-2); a mix of charcoal, soil and mordenite (Channel-3). No substrate (Channel-4).

Table 1. Description of the constructed wetland systems of the experiment.

Characteristic Design	Channel -1	Channel -2	Channel- 3	Channel –4
	HSF+FWS	HSF+FWS	HSF+FWS	HBF
Dimension	10,0 m long x 0.4 m wide x 0.3m depth	10,0 m long x 0.4 m wide x 0.3m depth	10,0 m long x 0.4 m wide x 0.3m depth	10,0 m long x 0.4 m wide x 0.3m depth
Vegetation species	Salvia splends, Mary gold, Petunia, Mint, Catharantus roseus, Kreson, Seri, Impatiens sultanii, Geranium, Cosmos.	Salvia splends, Mary gold, Petunia, Mint, Catharantus roseus, Geranium Kreson, Seri, Impatiens sultanii, Cosmos.	Salvia splends, Mary gold, Petunia, Mint, Catharantus roseus, Geranium Kreson, Seri, Impatiens sultanii, Cosmos	Salvia splends, Mary gold, Petunia, Mint, Catharant roseus, Geranium Kreson, Seri, Impatiens sultanii.
Substrate	Gravel	Mordenite (zeolite group)	Charcoal, soil and mordenite	Water
Depth of Substrate	26cm	26cm	26cm	9cm
Water depth from substraum	-10	-10	-10	-10
Influent loading rate	$2\ m^3/m^2$/dav	$2\ m^3/m^2$/dav	$2\ m^3/m^2$/dav	$2\ m^3/m^2$/dav
Loading method	Continue	Continue	Continue	Continue
Retention time	20 Min	20 Min	20 Min	20 Min

Table 2. Parameters studied in the experiment and analytical methods.

Parameter	Symbol	Analytical Method
Ammonium	NH_4-N NO_3-N	Technicon Autoanalyzer AA II
Nitrate	NO_2-N	Technicon Autoanalyzer AA II after cadmium reduction method
Nitrite	NO_3-N	Technicon Autoanalyzer AA II after sulphanilamid and N-(1-naphthyl) ethylendiamine
Total Nitrogen	TN	Technicon Autoanalyzer AA II after alkaline peroxydisulfate digestion
Dissolved Total Nitrogen	DTN	Technicon Autoanalyzer AA II after alkaline peroxydisulfate digestion
Total Phosphorus	TP	Technicon Autoanalyzer AA II after alkaline peroxydisulfate digestion
Dissolved Total Phosphorus	DTP	Technicon Autoanalyzer AA II after alkaline peroxydisulfate digestion
Phosphates	PO_4-P	Technicon Autoanalyzer AA IIafter ascorbic acid method
Biological Oxygen Demand	BOD	5-day BOD
Chemical Oxygen Demand	COD	Titrimetric method with K_2MnO_4
Suspended Solids	SS	GF filter gravimetric

Table 3. Minimum and maximum values of water temperature, pH, DO, conductivity and salinity from August to November of 1999 and from April to June of 2000. CSZ: a mix of charcoal, soil and mordenite substrate.

Month Year	Inlet	Channel-1 Gravel	Channel-2 Zeolite	Channel-3 CSZ	Channel-4 Water
Temperature (°C)					
Aug-99	27.9-29.6	28-29.2	28.3-29.4	28.4-30.2	29-30.5
Sep-99	25.8-29.1	25.3-28.4	25.1-28	24.9-29.6	25.3-29.1
Oct-99	19-24	14.7-23	15.1-22.8	15.2-22.7	13.6-23
Nov-99	15.1-17	10.1-15.6	11-13.6	11-14.7	9.4-13.5
Apr-00	14.5-16	11.5-15.9	12.6-16	13.5-15.7	12.5-16.9
May-00	19-22.4	16.8-22.8	17.2-19.6	17.6-22.4	17.4-23.1
Jun-00	21.8-22.3	21.9-23.1	21.7-23.4	21.4-23.7	21.9-23.1
pH					
Aug-99	6.9-7.3	6.8-7.2	6.8-7	6.9-7.8	6.9-7.6
Sep-99	6.9-7	6.9-7.2	6.8-7.1	6.9-7.5	6.9-7.2
Oct-99	7-7.3	7-7.3	6.9-7	7-8	6.9-7.3
Nov-99	6.4-7.3	7-8	6.7-7.5	7-8	7-7.4
Apr-00	7.1-7.2	7.4-8.1	7.1-7.4	7.5-7.7	7.6-7.8
May-00	7-7.3	7.3-7.4	6.8-6.8	7-7.1	6.9-7.2
Jun-00	7.2-6.7	6.9-7.3	6.7-7.2	7.3-7.4	7-8.6
DO (mg/L)					
Aug-99	7.9-8.2	7.7-8	7.8-8.1	7.8-8.1	7.8-8.5
Sep-99	8.4-8.6	7.8-9	8.2-8.9	7.9-9	8.2-8.6
Oct-99	87-9.9	9.2-11.5	9.4-11.2	9.3-11.2	8.8-12.1
Nov-99	8.3-11.1	11.1-11.8	10.4-12.8	10.5-11.7	11.5-12.7
Apr-00	10.2-11.1	10.2-11.6	10.4-11.4	10.5-11.1	10.7-11.7
May-00	8.9-9.7	9.5-10	9.4-10.1	9-9.4	8.8-9.6
Jun-00	8.8-9.5	8.9-9.4	8.9	9	9.3-8.7
Conductivity (mS/cm)					
Aug-99	15.3-24.4	9.2-24.4	16-24.7	15.3-24.6	15-24.8
Sep-99	7.21-11.5	7.3-11.5	7.3-11.5	7.2-11.7	7.3-11.5
Oct-99	11.6-20.9	10.1-22.5	7.7-21.9	11-22.1	10.7-22.1
Nov-99	1.9-6.3	1.9-6.3	1.8-6.3	2-6.3	2.1-6.3
Apr-00	0.6-1.8	0.2-2.7	0.3-2	0.4-1.9	0.3-1.9
May-00	6.8-8.5	6.8-8.6	6.8-10.6	6.8-10.8	6.8-10.9
Jun-00	4.7-7.1	4.8-7	4.8-7	4.2-7.2	4.8-7.1
Salinity					
Aug-99	0.5-1.5	0.5-1.2	0.5-1.5	0.5-1.5	0.5-1.5
Sep-99	0.4-0.7	0.4-0.7	0.4-0.7	0.4-0.7	0.4-0.7
Oct-99	0.7-1.3	0.6-1.3	0.4-1.3	0.6-1.3	0.6-1.3
Nov-99	0.1-0.3	0-0.3	0.1-0.3	0.1-0.3	0.1-0.3
Apr-00	0-0.1	0-0.1	0-0.1	0-0.1	0-0.1
May-00	0.4-0.5	0.4-0.5	0.4-0.6	0.4-0.6	0.4-0.6
Jun-00	0.2-0.4	0.2-0.4	0.2-0.4	0.2-0.4	0.2-0.4

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Table 4. Chemical data from 1999 to 2000 corresponding to Ammonium (NH_4-N), Nitrate (NO_3-N), Nitrite (NO_2-N), Total Nitrogen (TN), Total Phosphorus (TP), Phosphates (PO_4-P), Total Biochemical Oxygen Demand (TBOD), and Suspended Solids (SS), expressed in mg/L. In brackets is shown the percentage of removal in each one of the treatments. CSZ: a mix of charcoal, soil and mordenite substrate.

Month Year	Channel-1 Gravel	Channel-2 Zeoite	Channel-3 CSZ	Channel-4 Water	Month Year	Channel-1 Gravel	Channel-2 Zeoite	Channel-3 CSZ	Channel-4 Water
		NH_4-N					TP		
Aug-99	1145 (-29)	1060 (-2)	933 (-15)	1078 (-23)	Aug-99	257 (41)	261 (39)	168 (56)	365 (19)
Sep-99	567 (12)	444 (44)	798 (7)	566 (22)	Sep-99	256 (40)	323 (24)	188 (55)	384 (7)
Oct-99	130 (69)	470 (9)	203 (63)	484 (14)	Oct-99	140 (46)	248 (9)	134 (50)	296 (-11)
Nov-99	158 (66)	52 (87)	167 (60)	164 (53)	Nov-99	551 (-150)	723 (-194)	487 (-99)	798 (-204)
Apr-00	112 (84)	294 (58)	384 (43)	423 (36)	Apr-00	208 (22)	264 (0)	267 (-10)	231 (-2)
May-00	565 (42)	55 (94)	238 (85)	566 (42)	May-00	1338 (-240)	505 (-28)	115 (71)	437 (-11)
Jun-00	247 (70)	336 (65)	170 (83)	497 (41)	Jun-00	238 (56)	333 (41)	257 (53)	427 (26)
		NO_3-N					PO_4-P		
Aug-99	446	495	421	340	Aug-99	191 (-14)	181 (4)	133 (11)	206 (8)
Sep-99	210	191	311	137	Sep-99	151 (28)	254 (-20)	120 (43)	219 (-3)
Oct-99	7	5	3	76	Oct-99	115 (15)	151 (26)	103 (27)	237 (-71)
Nov-99	13	13	30	36	Nov-99	311 (-280)	442 (-394)	163 (-65)	237 (-63)
Apr-00	310	1216	622	263	Apr-00	161 (-12)	237 (-67)	163 (-17)	138 (0)
May-00	66	314	469	212	May-00	117 (-20)	75 (23)	71 (26)	254 (-134)
Jun-00	429	438	373	306	Jun-00	130 (26)	166 (12)	128 (28)	287 (-50)
		NO_2-N					TBOD		
Aug-99	47	95	95	129	Aug-99	4 (26)	3 (36)	4 (22)	5 (4)
Sep-99	17	16	20	195	Sep-99	4 (35)	4 (28)	3 (46)	5 (17)
Oct-99	7	5	4	59	Oct-99	2 (48)	2 (39)	1 (73)	2 (41)
Nov-99	5	5	19	21	Nov-99	3 (36)	2 (51)	3 (36)	3 (36)
Apr-00	1507	1195	697	492	Apr-00	6.7 (34)	7.8 (24)	10 (5)	9 (17)
May-00	48	272	268	105	May-00	2 (30)	1.1 (63)	1.5 (49)	3 (-16)
Jun-00	363	354	290	272	Jun-00	2.3 (35)	2.5 (30)	2.1 (42)	3 (21)
		TN					SS		
Aug-99	867 (29)	939 (22)	896 (25)	1056 (15)	Aug-99	8 (29)	6 (38)	9 (25)	9 (14)
Sep-99	553 (40)	540 (39)	508 (46)	660 (30)	Sep-99	10 (49)	10 (48)	12 (37)	14 (33)
Oct-99	528 (48)	641 (31)	467 (54)	931 (7)	Oct-99	6 (70)	7 (66)	6 (73)	9 (57)
Nov-99	728 (27)	632 (36)	657 (38)	723 (32)	Nov-99	8 (87)	3 (89)	14 (51)	6 (84)
Apr-00	1764 (12)	1460 (21)	1394 (29)	1431 (29)	Apr-00	6 (88)	6 (82)	10 (77)	6 (81)
May-00	908 (53)	737 (63)	1240 (34)	1292 (31)	May-00	3 (22)	3 (34)	3 (34)	10 (-135)
Jun-00	1380 (29)	1479 (24)	976 (50)	1307 (33)	Jun-00	26 (2)	24 (10)	17 (40)	21 (21)

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