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Original Research Article

Phytoremediation of industrial waste leachates by planted filters of *Phragmites australis (Cav)* Trin ex Steud*, Typha latifolia L.* and *Cyperus papyrus L*.

ABSTRACT : Industrial wastewater discharged into the environment without prior treatment promotes degradation of surface water and impacting groundwater quality. The aim of this study is to evaluate the purifying ability of a plant filter composed of three plants (*Phragmites Australis(Cav) Trin ex Steud,Typha Latifolia L., cyperus papyrus L.*) on leachates from a landfill of industrial waste. In order to assess purifying ability, we sampled 14 samples over 8 weeks, with 7 samples of the raw leachates from the technical landfill center, and 7 samples of leachates cleaned after passing through the tryptic vegetable filter. The physico-chemical analyzes made it possible to determine the following parameters: TOC, COD, NO_3^-, PO_4^{2-}, Ni, Cd, Cr^{VI}, Zn, Cu and Pb. The study of the purification performance of the system showed a significant decrease in organic pollution with abatement rates in TOC and COD greater than 90%. The average removal efficiency is respectively 45.97% for nitrates and 40.2% for phosphates. The abatement rates for heavy metals range from 41.2 % to 60.9 % for nickel, from 52.2 % to 68.5 % for cadmium, from 49% to 71.7 % for chromium VI, from 59 % to 74.6 % for zinc, from 50.9 % to 65 % for copper and from 61.4 % to 75.1 % for lead.

Keywords : Leachates, phytoremediation, phragmites autralis, Typha latifolia, cyperus papyrus.

1. INTRODUCTION

Problems of water pollution by untreated effluents have become a growing concern for developing countries. Increase of wastewater discharges that have not been adequately treated coming from industrial development, intensification of agriculture and increased volumes of wastewater related to rapidly urbanized areas, further promote degradation of surface water and groundwater quality worldwide [1]. One of the major challenges for wastewater in Africa is the general lack of infrastructure for collection and collection and treatment, which generates physical, chemical and biological pollution of the surface water in which it is discharged. Toxicity, mobility and the load of pollutants have a significant impact on water resources, human health and the environment [2]. According to the environmental audit report of the Congolese coastline on the coast of the city of Pointe-Noire, economic capital of the Republic of Congo, sewage from industrial, hotel and hospital establishments are rejected without prior treatment in the natural environment, especially in the sewers and rivers Tchinouka and Songolo [3].

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Recognized as a major environmental problem, the preservation of water resources is the object of intensive research. For two decades, the evolution of depollution have been witnessing techniques with the gradual transition from so-called traditional techniques to biodegradation techniques. Traditional depollution techniques highlight the complexity of the processes which, combined with the complexity of the pollution themselves, leads to the deployment of important logistical means for often high costs. Through this complexity are added the environmental requirements issued by local users who require not only the clean-up of contaminated environments but also a sustainable and perennial reconversion. Thus, to cope with the high costs of depollution and meet social requirements, professionals in the treatment market are moving more and more towards natural techniques that are less cumbersome and cheaper, by generating little waste and respectful of the environment to clean up and restore sites and polluted waters. Several natural processes related to plant growth can help clean up organic compounds, metals and even radioelements that contaminate sites and waters [4]. The resulting plant biotechnologies are encompassed under the generic name of phytoremediation [5]. Phytoremediation is a plant biotechnology based on the ability of plants to extract or block pollutants through the microorganism's activities that are present in the root environment (rhizosphere) or in the plant after absorption of the compound and degradation in the plant cells by specific enzymes, both in porous media, liquid or gaseous. It is therefore a biotechnology based on the synergistic cooperation of plant roots and soil microorganisms to decompose, transfer, deactivate and immobilize environmental contaminants [6; 7]. The main advantage of this process is to allow a pleasant transformation of the landscape with the installation of a plant cover (regreening, flowering) while depolluting the wastewater. It is used for the treatment of organic and inorganic pollution. It is therefore an interesting alternative to other physico-chemical depollution processes for wastewater before discharge into rivers because of their reduced environmental impact and low cost [8]. Previous studies have been shown that some plants and microorganisms can develop spontaneously in highly polluted environments [9]. This is the case of macrophyte lagoon stations (Typha, reeds) used to purify water or agronomic soils whose polluting load is reduced by an appropriate vegetation cover [10]. Macrophytes represent real water pumps whose water flow passes through the plant from the roots to the leaves. Phytoremediation has proved successful in the decontamination of waters polluted by heavy metals (copper, mercury, zinc, cadmium, iron, lead, etc.) [11; 12; 13]. It is based on growing plants with accumulative and tolerant properties for heavy metals. These accumulating plants are capable of extracting and accumulating heavy metals in their harvestable parts [14]. There are about 400 species recognized as hyperaccumulative and tolerant plants to heavy metals. These include sunflower, dandelion, rapeseed, barley, nettle and poplar [15]. For non-biodegradable organic pollutants, such as dioxins and furans, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) or pesticides, plants can be of great help. They can degrade organic pollutants in their cells by absorbing them or turning them into less toxic compounds [16]. The effectiveness of phytoremediation depends not only on the activity of plants, but also on the physicochemical properties of pollutants, their mechanisms of action, bioavailability and climatic factors [10]. The aim of this work is to evaluate the ability of plants such as common reed (*Phragmites austris (Cav) Trin. Ex Steud*), cattail (*Typha latifolia L.*) and papyrus (*Cyperus papyrus L.*) to eliminate or reduce the content of organic and inorganic pollutants in leachate from a landfill site.

2. MATERIALS AND METHODS

2.1 Presentation of the study site

Located about 35km north-west of the city of Pointe-Noire, in the village of TCHISSANGA (in the sub-prefecture of Loango), Loango Environment Society (SLE) is specialized in the management and treatment of industrial wastes in Republic of Congo. With an operating site covering an area of 22 hectares, the Loango Environment Society has a technical landfill site for the treatment of ordinary industrial waste, contaminated industrial waste and hazardous waste. This landfill site includes: a Technical Landfill (TL) for ordinary industrial waste (OIW) and a Technical Landfill (TL) for hazardous and soiled industrial waste (SIW).

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2.2 Alveoli structuration in the technical landfill

The alveoli of the technical landfill are totally sealed to prevent any groundwater pollution thanks to two types of safety (active and passive). The watertightness of the bottom and walls of the cells is obtained with natural materials, according to very strict impermeability standards. It is reinforced by a geo-membrane and a protective geotextile. This treatment makes it possible to control the emissions of pollutants by leaching thanks to the seals of the big-bags and the storage bin. The sealed alveoli of the technical landfill center consist of 5 superimposed layers and comprising: an anti-contaminant geotextile of 100 g/ m^2 in direct contact with the big-bags containing the waste, materials draining on 20 cm, a geotextile, a 10.5 mm thick PEHD geo-membrane and a GSB benthonic seal. The technical landfill is equipped with a network of drains to evacuate the leachate percolated into the massive waste to settling ponds.

2.3 Vegetal material

Three plants were used in the phytoremediation process: *Phragmite australis (Cav) Trin ex Steud, Typha latifolia L*. and *Cyprus papyrus L*. These plants were chosen for their ease of adaptation to local climatic conditions, their growth rate and their purification efficiency.

Common Reeds or *Phragmites australis (Cav) Trin ex Steud*: cosmopolitan species of perennial herbaceous plant of the family Poaceae, subfamily of Arundinoideae. The *Phragmites australis (Cav) Trin ex Steud*, whose rhizomes are very creeping, produce high robust stems (from 60 cm to two meters), robust and shiny. The common reed is the most widely used by filtering plant in the world to clean up wastewater whether it is in the form of a planted alluvial filter or in the form of planted basins. Carrying pure oxygen in its rhizomes, it is very efficient in treating organic COD, BOD_5 or TSS [17]. Basic plant for phyto-purification, it is installed at a water depth of 0 to 60 cm (distance between the rhizome and the surface of the water). Common reeds are tolerant to dry and wet conditions with a leachates pH between 3.7 and 9.0. In our study, the level of leachate to be treated is 7 to 10 cm.

Cattail or *Typha latifolia L.*: Cosmopolitan plant of the family of Typhaceae. *Typha latifolia* is an invasive and perennial 1-2 meter plant with a robust stem and broadly linear leaves (6 to 25 mm). It enjoys sunny areas. It settles quickly in a soil saturated with water or immersed to a depth of less than 30 cm (distance between the rhizome and the surface of the water). With the *Phragmites australis (Cav) Trin ex Steud, Typha latifolia L.* is the plant most used in phyto-purification (lagoons). *Typha latifolia* is a very resistant plant that can be used for the most desperate cases of pollution. It is able to clean up highly polluted wastewater at the limit of asphyxiation (liquid manure, waste water). It is very efficient in environments at the limit of anoxia (little oxygen). It can biodegrade petroleum products, chlorinated compounds and resists everything: heavy metals, salts, excess COD and BOD_5 [17]. Its roots stabilize substrates and allow suspended solids, carbon, nutrients and trace elements to integrate plant tissues. Cattails aerate the substrate by supplying oxygen between its stem and roots, which are points of attachment for microorganisms and provide nests for the development and feeding of microorganisms [18]. In our study, cattails are subjected to leachates with pH between 3.0 and 8.5.

Papyrus or *Cyperus papyrus L.*: Cyperus *papyrus L.* is a perennial herbaceous plant of the Cyperaceae family, native to the banks of the Nile and its delta. It forms large stands of reed marsh vegetation in shallow water. It can reach 2 to 3 m in height. Papyrus can be found in tropical forests, tolerating annual temperatures of 20 to 30 ° C and pH values of 5.5 to 8.0. Cattail prefers full sun to shady conditions. The use of *Cyperus papyrus L.* for the treatment of heavy metals in wastewater has shown a significant capacity in the retention of special metals for Cu, Zn and Iron [19].

2.4 Operational process of leachate Treatment

When stored in the technical landfill and under the combined action of rainwater and natural fermentation, the waste produces a liquid fraction called "leachate". Rich in organic matter and trace elements, these leachates are collected and purified before being released into the nature. The waste leachates from SIW technical landfill are first stored in a settling pond (Fig. 1) independent of the leachate coming from OIW technical landfill, in order to undergo a

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pretreatment. The OIW leachates are stored in another buffer tank.

Pretreatment of SIW leachates: The pretreatment aims to allow the decantation of suspended solids, the separation of hydrocarbons (oils, greases ....) and the aeration of volatile molecules such as H_2S present in the leachates. After decantation, the leachates pass through a hydrocarbon separator. Then pretreated leachates have been sent in the storage tank containing the leachate mixture coming from the two technical landfill (SIW and OIW).

Planted filters (PF): These are planted filters with horizontal flow (Fig. 2) whose base consists of gravel. They are composed of 3 parallel planted filters nominated as PF1, PF2 and PF3 (Fig. 3). Those 3 parallel planted filters are composed of common reeds, cattails and papyrus. Common reeds represent the majority portion of these horizontal planted filters. These planted filters have a horizontal sub-surface flow feed system and are continuously fed. The influents at the entrance are distributed using 3 channels which feed the 3 basins PF1, PF2 and PF3 allowing the lateral distribution of the flow. The evacuation systems consist of a drainage pipe located at the bottom of the basins and then connected to a output port. The phytopurification ponds are impregnated with synthetic geomembranes to prevent contamination of the groundwater. The mixture of SIW and OIW leachates from the settling tanks are driven by a PVC pipe to a common pond. The leachates then pass into the 3 filter basins covered with plants. The leachates simply stay in these 3 open ponds, populated with aquatic plants (common reeds, cattails and papyrus). At the entrance of each planted filter, there is a valve to regulate the flow of leachate. Purification is carried out by plants and micro-organisms attached to their roots. The output of each filter is connected to the exit pond where purified leachates are stored.

2.5 Leachate sampling procedure

In this study, sampling was performed over a 2 month's period. We first took samples of the leachate mixture from the SIW and OIW technical landfills: 7 samples of raw leachates were thus collected. Then, the raw leachates were sent to the 3 ponds of planted filters PF1, PF2, PF3. After an adaptation period of 2 months, the treated leachates are sent to the outlet storage tank: 7 samples of treated leachates were thus collected in the exit pond. Leachate samples were taken using a bailer (manual sampler) in the form of a plastic cylinder with a capacity of 1 L, provided at its lower end with a ball of no return. All samples are packaged in glass vials and stored in a refrigerator at 4 ° C and in the dark until analysis.

2.6 Leachate analytical methods

The pH and temperature were measured in the field to avoid variations, thanks to a EUTECH brand pH-meter and a TFX 420 brand thermometer. Total organic carbon (TOC) was measured from a test reagent heated for 2 hours in a CR2200 WTW thermo-reactor, and then measured using a S12WTW spectrophotometer. The chemical oxygen demand (COD) was

Fig.1. Settling pond of SIW leachates

Fig.2. Planted filters

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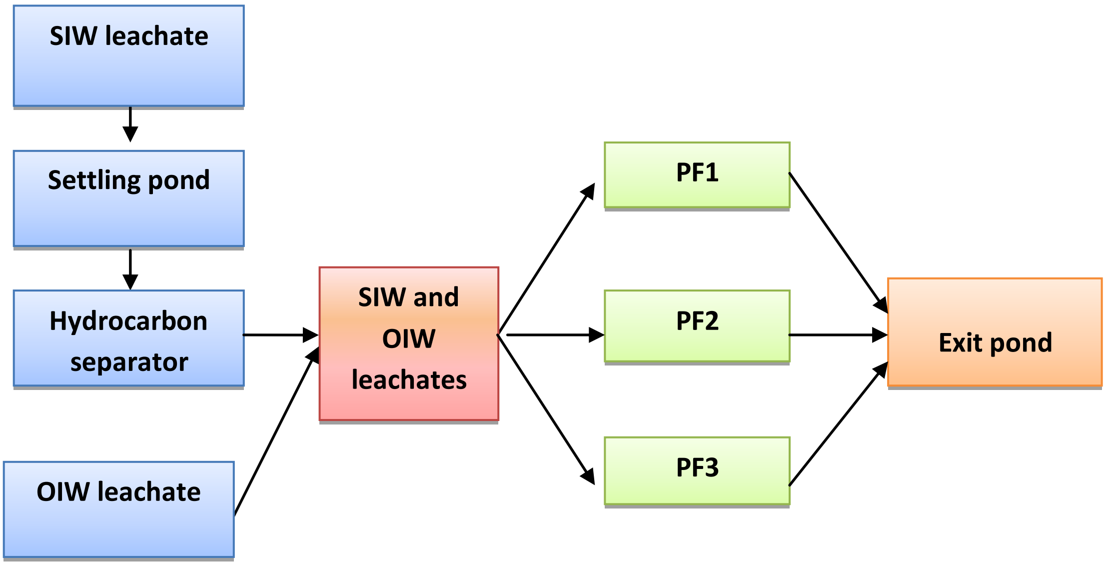

Fig.3. Scheme of the leachates purification plant

determined from the potassium dichromate method ($K_2Cr_2O_7$) and read directly from the S12WTW spectrophotometer. The trace elements, Cr, Ni, Cu, Zn, Cd, Pb, as well as the nitrate (NO_3^-) and phosphate (PO_4^{3-}) ions were dosed using the tube test reagents and measured at the same time using a S12WTW spectrophotometer.

RESULTS AND DISCUSSION

3.1 In situ parameters

Temperature: Leachates temperature values oscillate between 28.5 °C and 30.5 °C before treatment and between 29.7 °C and 30.2 °C after treatment (Table 1). The mean temperatures of influent and effluent leacheates are respectively 29.7 °C and 30 °C. These open basins justify that influent and effluent leachates temperatures are closely linked to the air temperature [20].

pH: Analysis of this parameter show that the pH values was between 6.51 and 7.03 before treatment and between 7,18 and 7,93 °C after treatment (Table 1). Influent pH values are well within the optimal range for bacterial growth which oscillates between 6.5 and 7.5 [21]. Effluent pH values show a general tendency of leachates to tend toward neutrality.

3.2 Efficiency of reeds planted filters

Physicochemical parameters: A significant decrease in TOC, DOC, nitrate and phosphate content in effluent leachates can be observed in comparison to influent leachates (Table 2).

Table 1. Influent and effluent leachates temperature and pH values

	T (°C)		pH	
Samples	**Influent**	**Effluent**	**Influent**	**Effluent**
S1	29.2	30.1	6.66	7.72
S2	29	30	6.55	7.66
S3	30.5	29.8	7.02	7.85
S4	30.1	30.4	6.75	7.78
S5	30.4	30.2	6.51	7.29
S6	28.5	29.7	7.03	7.93
S7	30.5	29.9	7.02	7.18

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Table 2. TOC, COD, NO_3^- and PO_4^{3-} values in leachates before and after phytoremediation treatment

Parameters	Influent leachates	Effluent leachates	Removal rate (%)
TOC-Total organic carbon (mg/l)	222-530	12-39	92.6-94.5
COD-Chemical oxygen demand (mg/l)	298-550	25-51	90.7-91.6
NO_3^--Nitrates (mg/l)	3-6.4	0.8-3.2	51.6-73,3
PO_4^{3-}-Phosphates (mg/l)	1-2.1	0.3-1.1	47.6-70

Fig. 4, Fig. 5, Fig. 6 and Fig. 7 show the variation of the TOC, COD, nitrates and phosphates of the effluents treated at the outlet of reeds plant filters compared to influent leachates. Fig. 4 shows a significant TOC reduction efficiency of 90.7 % to 91.6 % (Table 2). Fig. 5 also shows a significant difference in COD between influent and effluent leachates. In fact, the results show a reduction from 92.6 % to 94.4 % (Table 2) in effluents treated with reeds plant filters. These results are confirmed by Bensmina et al. [22] who obtained a COD purification performance of 66 % with *Phragmite australis (Cav) Trin ex Steud* and 79 % with *Typha latofilia L.* Daniel [23] observed an elimination efficiency of 80 % in COD in implanted basins of *Phragmites australis (Cav) Trin ex Steud* and *Typha latofilia L.* as well as Tiglyene et al. [24] that achieved an average COD removal of 74 % for the planted system. These good results with respect to organic pollutants can be explained by the fact that the planted mass allows a good elimination of the organic matter which is degraded by the bacterial activity at the level of the roots [25]. Also physical filtration mechanisms play an important role in the removal of TOC and COD because of organic solids can be collected in the gravel media. In Fig. 6 and Fig. 7, there is also a reduction in the content of nitrates and phosphates in the leachates from the filters planted compared to the raw leachates. The average removal efficiency ranges from 51.6 % to 73.3 % for nitrates and from 47.6 % to 70 % for phosphates (Table 2). The average removal efficiency is respectively 45.97% for nitrates and 40.2% for phosphates. These results are below of those obtained by Bensmina et al. [22] who observed a reduction in nitrates and phosphates, respectively of 62 % and 59% and those of Saloua et al. [26] who obtained a reduction rate of 49 % in phosphates. These reductions can be explained by the fact that nitrates and phosphates are used as nutrients by plants for their tissues growth [27], but also because they can be stored in roots, rhizomes, stems and leaves [28].

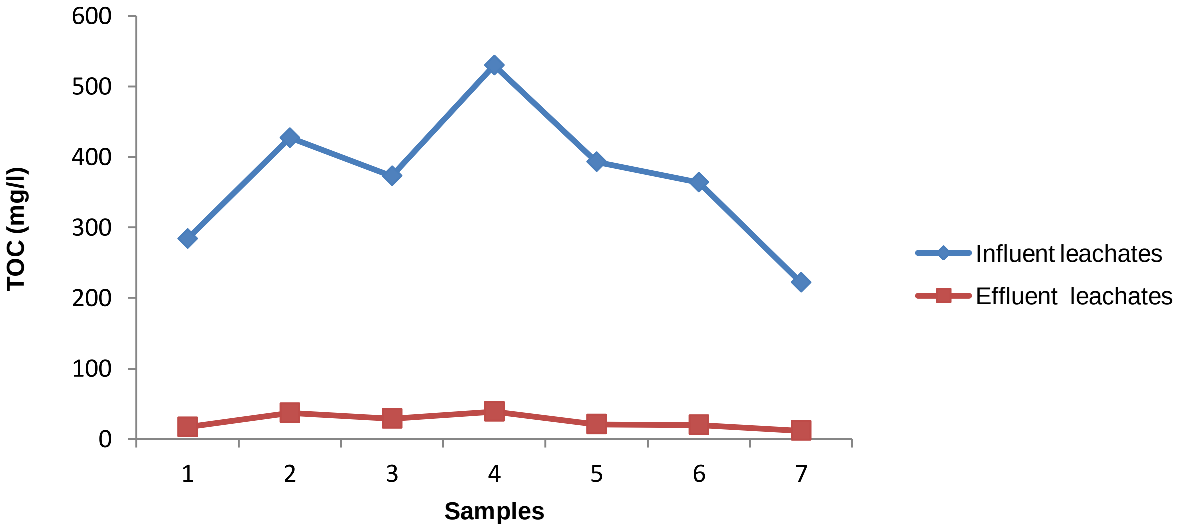

Fig. 4. TOC variations in leachates before (influent) and after (effluent) passage through reeds planted filters

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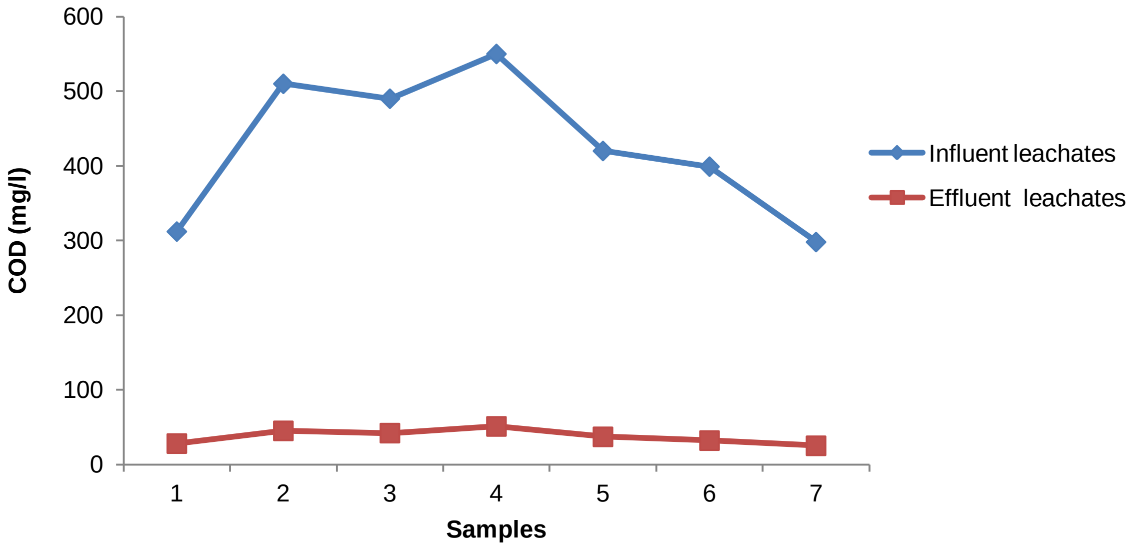

Fig. 5. COD variations in leachates before (influent) and after (effluent) passage through reeds planted filters

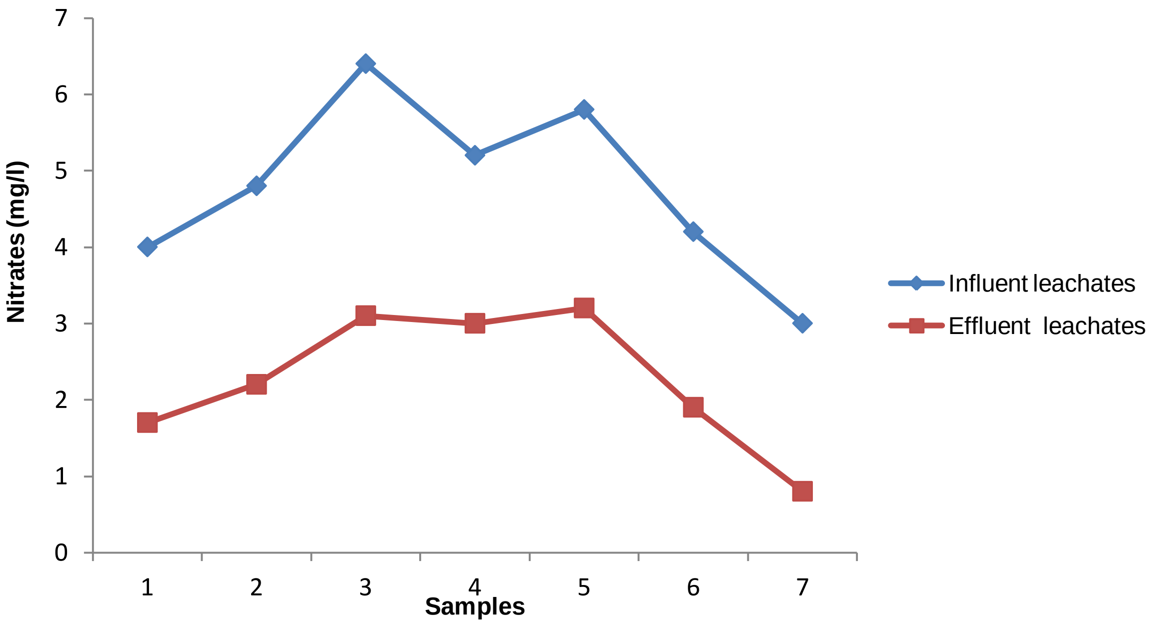

Fig. 6. Nitrates variations in leachates before (influent) and after (effluent) passage through reeds planted filters

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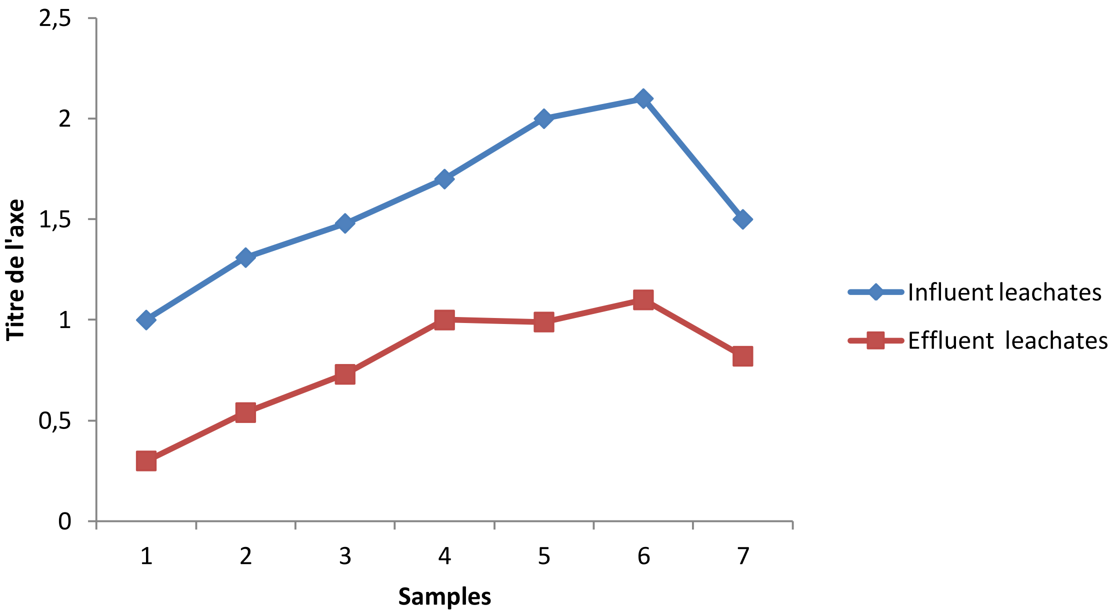

Fig. 7. Phosphates variations in leachates before (influent) and after (effluent) passage through reeds planted filters

Heavy metals parameters: The range of the heavy metals leachates composition before and after treatment and the removal rate in wastewater are collected in table 3. The range of heavy metal abatement is found to be between 41.2% and 75.1 % in leachates from planted filters compared to raw leachates. The average removal efficiency ranges from 41.2 % to 60.9 % for nickel, from 52.2 % to 68.5% for cadmium, from 49% to 71.7% for chromium VI, from 59 % to 74.6 % for zinc, from 50.9 % to 65 % for copper and from 61.4 % to 75.1 % for lead (Table 3). These results are below those observed by Menka Kumari et al [29] who obtained a range of reductions between 60 % and 80 % for heavy metals (Ni, Cd, Cr, Zn, Cu and Pb) contained in wastewater using a combination of plants consisting of *Phragmites australis (Cav) Trin ex Steud* and *Typha latifolia* L. Although the physicochemical analyzes of the different parts of the plants were not carried out in this study, these results may be due to the accumulation of heavy metals in plants as many studies have shown: Klink [30] studied the concentrations of Zn, Cu, Cd, Pb and Ni in the roots, rhizomes, stems and leaves of both species (*Phragmites australis (Cav) Trin ex Steud and Typha latifolia L.*) in polluted waters, and showed that despite differences in trace metal accumulation capacity between species, concentrations of Cu, Zn, Pb and Ni in macrophytes had the following accumulation pattern: roots> rhizomes> leaves> stems. Fonkou et al. [31] results revealed an increase in Cd, Cu, Pb and Zn concentrations of water from the Olezoa wetland complex to *Cyperus papyrus L.* plants. Thus, numerous studies prove that the plants of *Phragmites autralis (Cav) Trin ex Steud, Typha latifolia L. and Cyprus papyrus* are hyperaccumulators that can be used for phytoremediation of wastewater [32, 33, 34].

Table 3. Heavy metals leachates composition (mg/l) before and after phytoremediation treatment and remove rate (%)

Parameters	Influent leachates	Effluent leachates	Removal rate (%)
Ni-Nickel (mg/l)	1.79-4	1.05-1.56	41.2-60.9
Cd-Cadnium (mg/l)	2.4-4.47	1.15-1.41	52.2-68.5
Cr(VI)-Chromium (mg/l)	0.86-1.66	0.44-0.47	49-71.7
Zn-Zinc (mg/l)	1.24-2.34	0.51-0.59	59-74.6
Cu-Copper (mg/l)	0.91-2.41	0.45-1.08	50.9-65
Pb-Lead (mg/l)	1.28-2.26	0.49-0.56	61.4-75.1

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3.3 Tolerance of reeds planted filters in the liquid environment

However plants showed an intolerance to the liquid environment since they disappear completely after 3 months (Fig. 8). This gradual disappearance of the plants may be due either to an excessive concentration of metals in the leachates to be treated, or to the salinity of the environment. Indeed, metals are essential for the good functioning of a living cell because they intervene in many functions [35]. However, the cell only needs these ions in small quantities. When the intracellular concentration of these elements becomes too great, the excess of metal reacts with O_2 to form free radicals [36]. However, these molecules are extremely reactive compounds capable of oxidizing biological molecules (proteins, DNA, lipids, ...) causing major damage and control its concentration in metal ion knowing that they are necessary but an excess is lethal.

In our study, the heavy metal content of leachates is less than 5 mg/l, that are not to trouble shooting for these hyperaccumulative plants that have systems allowing them to grow in a hostile environment [38]. In addition, the salinity measurements of the treated leachates reveal the presence of Nacl with high contents ranging from 3.5 g/l to 18.8 g/l (Fig. 9) and an average content of 12.28 g/l. The results obtained by Bounkala et al. [39] show that saline stress can create physiological and biochemical disturbances. Indeed, when reeds such as *Phragmites australis (Cav) Trin ex Steud* are brought into contact with saline waters with salinity higher than 10 g/l, no growth of plants occurs, that is to say that all plants die [40]. High concentrations of NaCl therefore decreased the productivity of the plants, which hampered their growth in the medium.

Fig. 8. Progressive disappearance of plants in planted filters: a) Planted filters at 1 day b) Planted filters at 2 months c) Planted filters at 3 months

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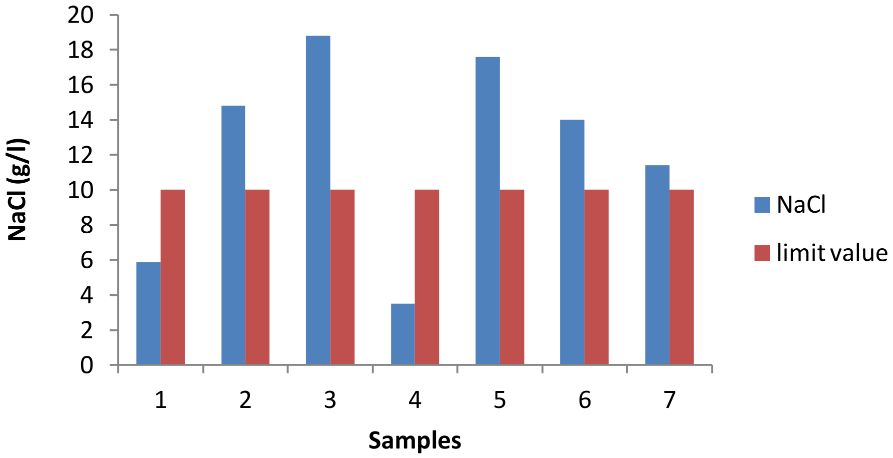

Fig. 9. NaCl content in effluents after passage in reeds planted filters

4. CONCLUSIONS

In this study, we evaluated the purifying abilities of a plant filter composed of three plants (*Phragmites Australis (Cav) Trin ex Steud, Typha Latifolia L.* and *Cyperus papyrus L.*) on leachates from a landfill of industrial waste. The results obtained showed a significant decrease of organic pollution with TOC and COD reduction rates over 90%. The reduction of nitrates, phosphates and heavy metals (Ni, Cd, Cr, Zn, Cu, Pb) in effluent leachates after passing through the reed plant filters was also highlighted, but their rate of abatement does not exceed 75%. These results confirm the performance of filters planted with *Phragmites Australis (Cav) Trin ex Steud, Typha Latifolia L.* and *Cyperus papyrus L.* even though the salinity related to the nature of the wastewater somewhat reduces their purification capacity over time. Treatment with macrophyte plant filters appears to be an effective alternative for the treatment of industrial wastewater. This work has got a promising future. An investigation could be very interesting to assess the relationship between bacteria and different plants. Bacteria could play a very important role in symbiotic abilities with plants in terms of cell-cell interactions.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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