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Evaluation of the efficiency of different materials to remove specific pollutants from landfill leachate

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ABSTRACT

The management of solid waste and its by-products is considered as one of the major concerns. Namely, the leachate and biogas that are collected, treated and transported directly to the landfill. For this reason, this study aims to characterize these Leachate samples and to evaluate the technical efficiency and the socio-economic performance of an infiltration-percolation treatment as a technique, which is adopted on appropriate fixed natural supports. In this regard, we evaluated the performance of the various adsorbents tested for the treatment of leachates, and then we analyzed the results of the best filtration media from the point of view of purification performance. The physical–chemical monitoring of these tests employs activated carbon filter and clay. However, the mixture of these two elements has shown that we obtained an average reduction in pollution in terms of suspended solids with about 80%, 78% and 94%, respectively, an average reduction of the chemical oxygen demand of 90%, 69% and 73%, respectively, but with an increase in conductivity and pH for the sand filter media.

Keywords: Infiltration-percolation; Chemical oxygen demand; Suspended solids; Leachate; Physical–chemical parameter

1. Introduction

Landfill leachate, as a highly complex polluted wastewater, is not only associated with the percolation of rainwater through solid waste, but also from biological, physical and chemical processes within the landfill [1]. The challenges of disposal treatment and, in general, the management of municipal waste in developing countries remain critical because of the population growth and socio-economic activities intensify. In developing countries, the most widely adopted method of solid and liquid waste management is landfilling [2,3], where all types of waste are untreated and mixed together [4].

The major and unavoidable consequences of landfilling are the production of leachate, which can impose several environmental problems [5,6]. Indeed, these leachates are rich in organic and inorganic matter, and heavy metals [7]. The physical–chemical composition of the leachates is specific to each discharge. It depends on certain parameters such as the nature, the age of the landfill, the

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type of waste, the method of disposal, the nature of the site, and the climatic conditions [8]. The objective of this paper is to study the physical and physical–chemical characterization of the leachate of the Tunisian landfill and to find out a less expensive and efficient process for the treatment of these leachates [9].

The process aims to obtain a very significant reduction in the organic leachate load that could be appropriate for similar cases which perfectly adapt to the socio-economic context. This contribution will therefore consist of a physicochemical treatment by infiltration-percolation batch wise of the polluting load. For this, nine types of adsorbents were tested in this study: sea sand (S), the core of dates (CD), clay soils (CS), sawdust (B), agricultural waste (AW), sea plant (SP), cactus (C), fruit of the caesarian tree (A), and activated carbon powder (ACP).

Infiltration-percolation is a physical process, generally disposed after decantation, for clarifying a liquid that contains suspended solids by passing it through a porous medium of granular material [10–12]. It is based on the adsorption phenomenon, which is defined as a collection of substance at the interface between the phases. It is a surface phenomenon that involves a solid adsorbent and a fluid, gaseous or liquid adsorb ate. It comes originally from intermolecular forces of attraction. Adsorption occurs when a molecule is attracted more strongly by the surface of one of the phases. The phase that adsorbs is called adsorbent and the molecules that adsorb are the absorbate [13–15]. Adsorption remains among the most used techniques and easy to implement. The elimination of dyes in aqueous solutions by adsorption on different solid materials, in particular on activated carbon, has been the subject of much work [5,11]. Adsorption of organic molecules such as dyes onto activated charcoal has proven to be a very effective treatment technique, however in the case of some recalcitrant dyes, charcoal overdoses are required for improved efficiency, making the cost of the excessive operation. In addition, the regeneration of activated carbon is a delicate operation and does not unanimously agree on its usefulness [16–18]. The added value of this study is the valorization of the local natural adsorbents considered as waste in the treatment of another waste which is leachate, by a simple, easy and eco-friendly process.

2. Material and methods

2.1. Physical–chemical characterization of leachates

2.1.1. Sampling

The leachate samples were carefully analyzed to obtain a homogenous and representative sample. Due to the lack of a leachate drainage system from the landfill, we sampled several well-defined drainage points. The volume taken for each sample is 1 L. These leachate samples are collected in polyethylene bottles, which are stored in a cooler, and transported directly to the laboratory for analysis.

2.1.2. Physical–chemical analyzes

Different types of analyzes were performed on sampled leachates. Some measurements are carried out immediately, at the time of sampling, to avoid changing the characteristics of the samples, as is the case with temperature (digital thermometer Type GTH 1160), electrical conductivity (conductivity meter HANNA EC214) and pH (pH-type meter, G BOYER, Jenco 6173). Others have been carried out in the laboratory: BOD_5, chemical oxygen demand (COD), suspended solids (SS), nitrate, nitrite, ammonium and chloride concentrations.

All chemical leachate parameters were determined using standard wastewater analysis methods [19,20]. The suspended solids content (SS) was determined by the centrifugation method. The COD is analyzed using a Behr TRS 300 thermo-reactor type DCO-meter.

Nitrite, nitrate and ammonium compositions were determined with a spectrophotometer in the visible type Portable Colorimeter Photoflex. In order to determine the rate of reduction of the treated leachate parameters, all the physical–chemical analysis tests were repeated. Thus the average values are determined according to the standard AFNOR techniques, which are summarized in Table 1.

2.2. Structures and morphology of the adsorbents used

2.2.1. Sea sand (S)

Sand is a granular material composed of small particles coming from the deterioration of materials of mineral roots (mostly shaking) or natural (shells…). In our study, we used sea sand from a Tunisian beach.

2.2.2. Core of dates (CD)

The date kernels were thoroughly washed with distilled water and then dried in an oven at 105°C for 24 h. They are then crushed and sieved to retain only the fraction between 0.5 and 2 mm [21].

2.2.3. Clay soils (CS)

In our study, we used natural clay soils, taken from the region of Sidi Thabet to the surrounding of INRAP.

Table 1
Physical–chemical characterization of raw leachates

Parameters	Values
pH	8.45
Temperature, °C	22
Turbidity, NTU	588
Suspended solids, mg/L	640
Salinity, g/L	45.6
Total dissolved solids, g/L	26.3
Conductivity, mS/cm	60.6
Nitrate, mg/L	170
Nitrite, mg/L	10
Chemical oxygen demand, mg/L	46,250
Phosphate, mg/L	490
Chloride, mg/L	10,700
Al, mg/L	350

The choice of these materials is justified by the following characteristics:

- Abundance of these soils in the Sidi Thabet region, with accessibility and low cost.
- Richness in very fine mineral constituents. These make them more active in solution, which gives them a high adsorbent capacity [22].

The clay soil removed was previously milled, sieved and dried at 105°C. This last operation makes it possible to eliminate the water of hydration, which makes the material hydrophobic.

In this regard, the clay soil is provided with extremely small clay particles, which are less than two microns. Therefore, this particular operation allows microorganisms to more easily ingest nutrients. The tiny size of the particles makes this earth less aerated and therefore more porous (Fig. 1c). A clay soil will take much longer to warm up, resulting in late blooms in some gardens [41].

From sieving of our soil, we can conclude that the predominant size of the clay soils used is 500 μm. This small particle size will facilitate the attachment of the organic material to the surface of the adsorbent material, which is generally greater when the particle size of the material is low. For this reason, we chose fractions with a particle size ≤500 μm to carry out the various leachate filtration tests.

2.2.4. Sawdust (B)

In this part of the study, we used two types of natural sawdust of the following species: Sawdust of *Cedrus atlantica* (B1), consisting of yellow-colored grains of coarse and loose texture in the dry state. Sawdust from the roots of *Arundo donax* L. (B2), consisting of whitish grains of coarse and loose texture in the dry state.

The sawdust or slash and wood processing residues are specifically studied as major biomasses whose adsorption efficiencies are confirmed for metal adsorption. This focused mainly on sawdust of the following species: red fir (*Abies magnifica*) [18], spruce (*Picea engelmannii*) [39], pine (*Pinus roxburghii*) and kamatsu (*Pinus densiflora*), as well as *Bassia latifolia* [40]. However, this research did not confirm the purification efficiency of these processes from the point of view of the organic pollutant load.

Red fir is a shrub from the Mediterranean region, which grows in rocky places, especially on limestone. It is an evergreen tree that rises to 40/60 m in height and whose trunk is about two meters in diameter. The Evergreen species longevity is greater than 500 y, in comparison with the cedar; the main vegetation cover of the Moroccan mountains.

2.2.5. Agricultural waste

Agricultural residues refer to the fibrous parts of cereals usually left in the fields. They have in common the fact of being made up of the parts of the plants not consumed by the man after the harvests, but also of having a low nutritional value for the animals and of having very little or no food value for them. Monogastric Yet one of their uses is in the diet of animals, despite everything capable of transforming them. In this study, we will limit our choice to agricultural residues commonly used agricultural residues of cereals.

Fig. 1. The visual appearance of sawdust 1 (a), the core of dates (b), fruits of the Casuarina tree (c) and sawdust 2 (d).

Fig. 2. The visual appearance of the cactus (a), the sand (b) and clay soils (c).

2.2.6. Sea plant (SP)

The algae plant organism is able to photosynthesize through chlorophylls (Fig. 3b), but it does not acquire advanced reproduction of higher plants (*Embryophytes* or *Archégoniates*: mosses, ferns, conifers, angiosperms) [23]. This definition remains vague and does not show that in fact there are more genetic distances between the various groups of algae than between the higher and the multicellular animals. We cannot even use their living environments in this definition [24].

2.2.7. Cactus (C)

The cactus is a tree, which natively came from the arid and semi-arid regions of Mexico. It belongs to the genus *Opuntia*, it is a succulent xerophytic plant capable of storing a large quantity of water and presenting no danger to human health [25]. It also has considerable values in the fields: cosmetic, medicinal and food [26,27].

In this study, we are proposing to test a bioadsorbent based on cactus (prickly pear), from the region of Sidi Bouzid of Tunisia. The cactus was collected and washed with deionized water. The washed cacti were dried in the sun for several hours until they completely lost the moisture content that was present. They were stored in a clean plastic box and dried for later use (Fig. 2a). The cactus powder was accurately weighed using an electronic scale.

The outer pad was considered the outer layer of light green tissue composed of chlorenchyma and the inner pad was considered the inner layer of off-white tissue composed of parenchyma. The inner pads were then cut into small pieces to facilitate drying. The sliced cactus was kept in the oven at 80°C for 8 h for drying. The dried cactus was ground into a fine powder using a mixer and sieved to sizes ranging from 53 to 106 μm [6].

2.2.8. Fruit of the Casuarina tree (A)

The origins of *Casuarina equisetifolia* tree went back to tropical Asia: Indonesia, Malaysia, India, Sri Lanka, northern Australia and Melanesia: New Guinea, New Caledonia, Fiji. This species have been introduced as an ornamental for its wood, to fight against erosion, in many tropical regions and has sometimes naturalized [28]. The *Casuarina equisetifolia* is easy to grow because this fast-growing tree likes the sunlight, high temperatures, and very low water requirements [29]. The fruits are woody and spherical cones 12 mm in diameter, persisting for a long time on the tree. At maturity, the cones reveal numerous small openings with two pointed scales each containing a seed [30].

2.2.9. Activated carbon powder

The carbon used, which is of plant origin, was first ground in a mortar and then analyzed by particle size in order to obtain fine particles, characterized by a diameter of between 0.08 and 0.1 mm ($0.08 \leq d < 0.1$). The activated carbon used has a high affinity for the two herbicides studied because of the small size of their molecules and the favorable interaction between these solutes and the material [31].

2.3. Infiltration percolation of leachates

In our study, we used twelve columns with one or more natural adsorbents (Fig. 4). In order to standardize the experiment parameters, we used the same conditions for all columns. Essentially, a flow rate and a uniform and constant contact surface, a particle size of: ≤500 μm, an average temperature of 22°C, and the preparation of all adsorbents at the same time and under the same conditions. Therefore, we tried to evaluate the organic matter and consequently the pollutant load of the samples studied according to physical–chemical parameters (pH, total dissolved solids, salinity, conductivity, COD, P, Cl, SS, NO_3^-, NO_2^-).

3. Results and discussion

3.1. Physical–chemical composition of leachates

The leachate characteristics are difficult to be determined because they evolve in time and space. Since these leachates are enriched over time in non-biodegradable compounds, conventional treatments do not always make it possible to meet discharge standards [32,33].

The leachates of this work are characterized by a high organic matter (high COD), but easily biodegradable. The suspended solids (SS) found in the leachates studied are of the order of 640 mg/L, this concentration decreases slowly as the site ages. They also give the leachate a cloudy appearance, a bad taste and a bad smell [12,34].

The pH recorded in the leachate from the studied landfill is around 8.45. The basic hydrogen potential characterizes

Fig. 3. (a) Agricultural waste, (b) sea plant, and (c) fruit of the Casuarina tree.

old discharges with aged or stabilized leachate. This proves that waste does not accumulate on the site. This is probably due to the mode of operation of this site, which is based on burning waste and intensive cattle rearing [35].

The leachates studied have a high electrical conductivity of the order of 60.6 mS/cm due to the contributions, by the discharge, of very mineralized leachate, under the effect of the cations and anions and very high value of the salinity, which favors bacterial development [36,37].

The concentrations of phosphates, nitrates and nitrites are 490, 170 and 10 mg/L respectively. In this type of discharge, nitrogenous matter (organic nitrogen and ammoniac nitrogen) is mainly reduced due to the urea of human or animal. The direct release of reduced nitrogen into the environment consumes oxygen and is detrimental to fish life [38].

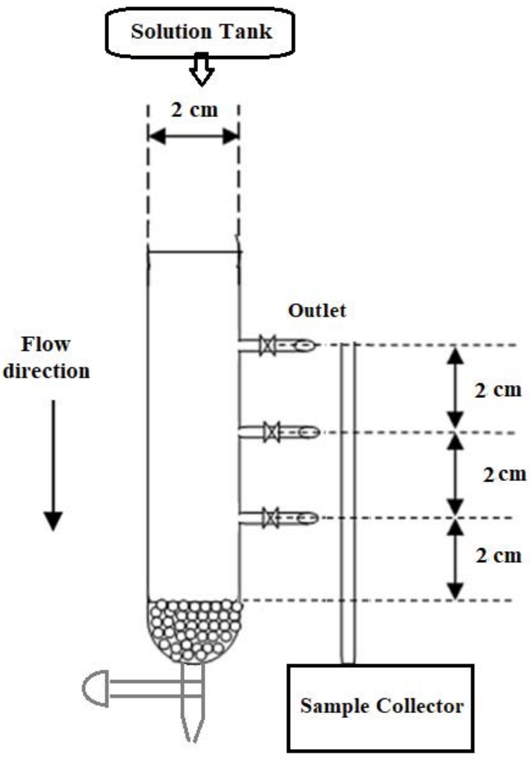

Fig. 4. Fixed bench support for experiments.

3.2. Adsorbent characterization before and after adsorption

In Fig. 5a we have the infrared spectrum of sawdust before and after adsorption. We find that the spectrum of sawdust before adsorption shows a broad band in the 3,600–3,200 cm^{-1} region which is due to the elongation of –OH bonds, while after adsorption we notice the absence of this band [18].

The thin band around 1,435 cm^{-1} in the spectrum after adsorption is attributed to vibrations of the C=O groups of carboxylic acids, ketones and aldehydes [39] (Fig. 6c). The band around 1,590 cm^{-1} corresponds to the vibration of the C=C groups of carboxylic acids and ketones (Fig. 6d). Thin band around 1,000 cm^{-1} characteristic of the C–C bond of the spectrum before adsorption [6,40]. The difference between the two spectra is due to the phenomenon of adsorption on the surface of cactus. The infrared spectrum shown in Fig. 5b shows that there is no large change in the absorption bands of cactus after and before adsorption, almost the same bands on both spectra [41].

The two infrared spectra of the cactus before and after adsorption (Fig. 6) showing that the adsorption has no effect on the structure, because the two spectra represent the same bands.

3.3. Results of infiltration-percolation leachate treatment tests

The leachates treated by the filtration columns (5), (9), (10) and (13) show a reduction of the organic pollution in terms of COD and P with a maximum of pollution for the filtration column (9). This gave a 96% reduction in COD content with 550 mg O_2/L, which is significantly lower than the standard (1,000 mg/L) and a 93.83% reduction in P content with a value of 9 mg/L, lower than the standard (10 mg/L). This maximum reduction in organic load is explained by the significant adsorption properties of finer minerals, particularly clay, activated carbon and silt, these clay minerals have a lamellar structure that allows the insertion of organic molecules between the leaflets.

3.3.1. Hydrogen potential (pH)

For natural adsorbents: sawdust, cactus, clay soils or dates, the pH does not mark a significant evolution (Fig. 6c). However, we noticed an increase in the leachate pH after filtration in the columns containing sands.

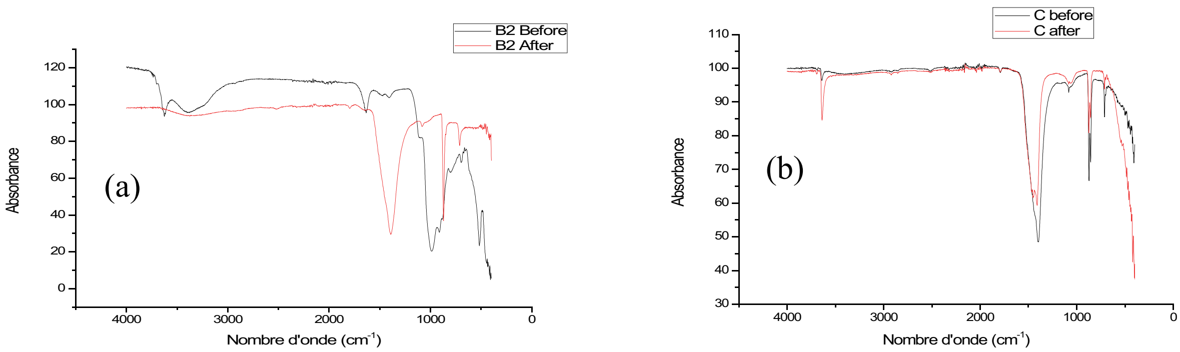

Fig. 5. Evolution of infrared bands before and after adsorption for sawdust (a) cactus (b).

Probably, this natural basicity, mainly from feldspars, which are alkaline aluminosilicates [42], corresponds to the release of ions of a basic nature in leachates [43,44].

3.3.2. Washing and transit time of Colon

The transit time is the average time required for an element to pass through a system at equilibrium. One way to determine this time is to calculate the time required to fill the system with the element under study.

After the use of the filter material, the filtration becomes ineffective, which requires washing the filter [45]. This washing is done by injecting clean water only. These injections are made at the top of the filter. The speed of the wash water is limited due to possible material losses.

The water-borne wash hangs a long time; the fruit filter wash of the Casuarina tree needs a long time in comparison to other supports (Fig. 6d).

3.3.3. Abatement of electrical conductivity, total dissolved solids and salinity

The evolution of the electrical conductivity during filtration by column shows decreasing values related to chemical exchanges between water and the filter material (precipitation and adsorption). This reduced the electrical conductivity of the filtrate to 92% as maximum reduction with the activated carbon column (Fig. 7) with a conductivity of the order of 2,400 µs cm^{-1} lower than the Tunisian standards for rejects.

Salinity refers to the amount of salts dissolved in a liquid, especially water, as a powerful solvent for many minerals. Salinity should not be confused with the hardness of water which is related to its calcium and magnesium dosage [46].

3.3.4. Abatement of COD and turbidity

Since sandy clay contains a high silica content, a large adsorbent with a high electrical polarity and mineral elements, particularly ferrous ions, the latter contribute to the neutralization of the negative charges of organic matter contained in the taper. However, the reductions in COD for the sawdust columns are in principle due to the free hydroxyl bonds available in the macromolecules of cellulose, hemicellulose and lignin (Fig. 10a). The elimination of the organic load by this type of the adsorbents is correlated with that of the leachate turbidity (Fig. 10b). This confirms that the suspension of leachates is produced by the organic matter involved [47].

3.3.5. Abatement of suspended matter and nitrite

The suspended solids occur in leachate in various forms: suspended solids, organic suspended solids and live suspended solids. Because of repulsive phenomenon, colloids

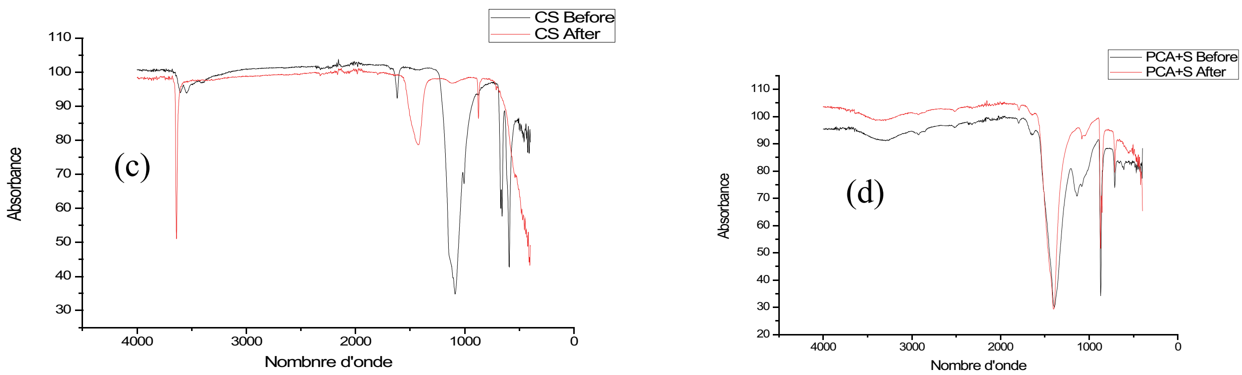

Fig. 6. Evolution of the infrared bands of clay soil (c) and activated carbon powder + sand and (d) before and after adsorption.

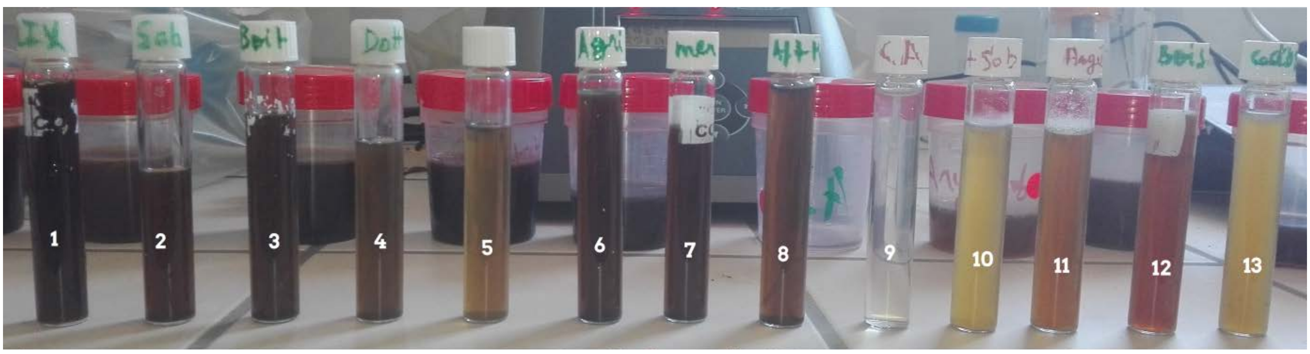

Fig. 7. Visual appearance of filtrates from different matrices: 1: leachate; 2: sand (S); 3: sawdust (B1); 4: core of dates (CD); 5: fruit of the Casuarina tree (A); 6: agricultural waste (AW); 7: sea plant (SP); 8: agricultural waste + sea plant; 9: activated carbon powder + sand; 10: clay soil (CS) + sand; 11: clay soil (CS); 12: sawdust (B2); 13: cactus (C).

generally form very stable suspensions (Fig. 11a). The adsorption phenomenon then act to neutralize their charges, to promote their agglomeration and allow their settling. The suspended matter will therefore be trapped in the granular mass of the filter and a form aggregate in the case of flocculation, which explains the results illustrated in Fig. 8a and shows a maximum reduction to 94% by filtration on the support (ACP + S) with a content of 210 mg/L (standard = 400 mg/L).

Since nitrite is a key nutrient in porous natural environments, its quantification has seen the development of several monitoring methods for different study environments: natural waters, saline waters, soil solutions [48] (Fig. 11b). Fig. 9b shows that the filter substrate (CS) contributes to a maximum reduction of nitrite ions with a respective reduction rate of 95.2%.

3.4. Modeling of leachate adsorption kinetics

In modeling the adsorption kinetics of leachate on the two supports, we were interested in pseudo-first-order and pseudo-second-order kinetic models.

3.4.1. Pseudo-first-order and second-order model for clay soil

The adsorption modeling is represented by the pseudo-first-order model and the pseudo-second-order model (Table 2), in Fig. 12.

We can notice a good linearity of the curve for the pseudo-second-order model (Fig. 12b), the adsorption on the clay soil follows well the kinetic model of pseudo-second-order with a correlation coefficient close to 1 ($R^2 = 0.996$). Moreover, we note that the calculated adsorbed quantity (56.21 mg/g) is close to that found experimentally (76.5 mg/g).

3.4.2. Pseudo-first and second-order model for cactus (C)

The adsorption modeling is represented by the pseudo-first-order model and the pseudo-second-order model, in Fig. 13.

In the same way as for the adsorption of leachate on clay soil, the adsorption on cactus follows the pseudo-second-order kinetic model with a correlation coefficient close to 1 ($R^2 = 0.995$). It can also be seen that the calculated adsorbed quantity (82.30 mg/g) (Fig. 14b) is quite close to the experimentally determined one (72.1 mg/g) (Fig. 14b), while the difference between the calculated adsorbed quantity and the experimentally determined one, for the pseudo-first-order model (Table 3) is much more significant [49].

3.5. Adsorption isotherms of leachate on the two supports

To understand the reaction mechanism involved, we used Langmuir and Freundlich isotherms (Table 4).

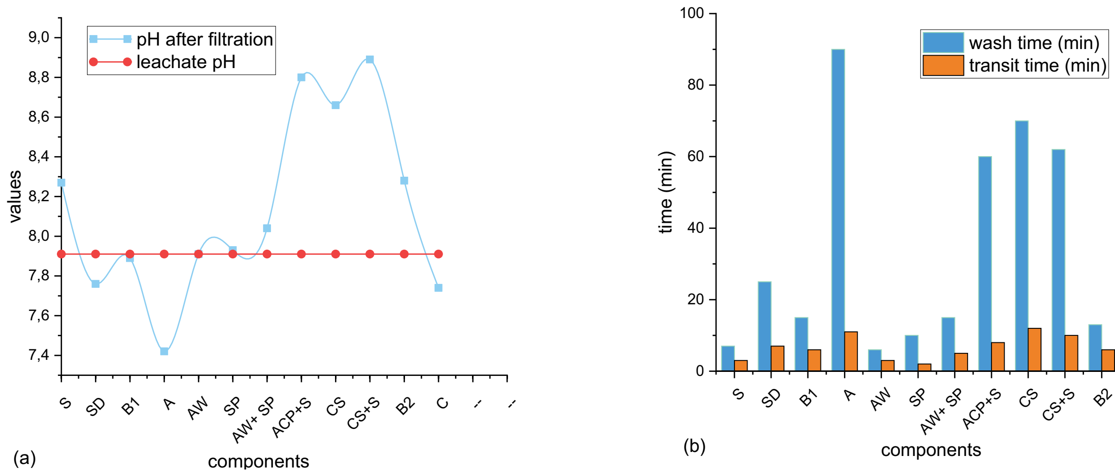

Fig. 8. (a) Evolution of the pH after filtration and (b) evolution of the transit time and filtration.

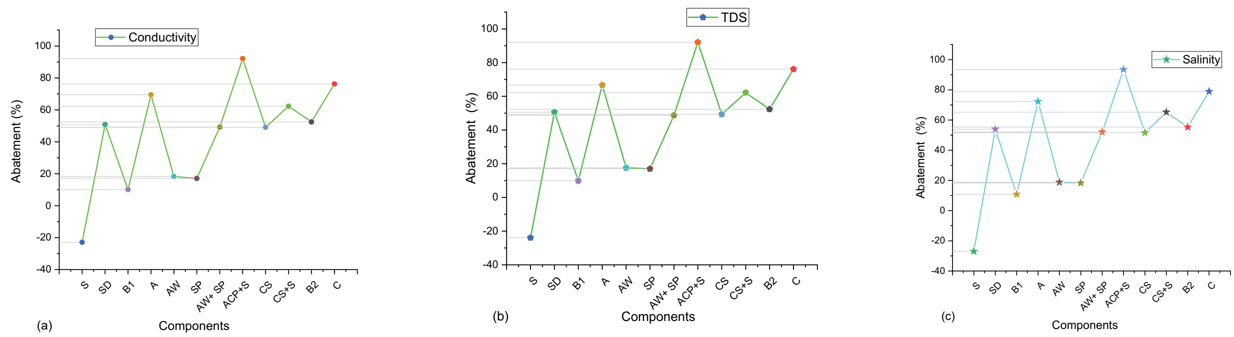

Fig. 9. (a) Conductivity abatement, (b) abatement of the total dissolved solids and (c) abatement of salinity.

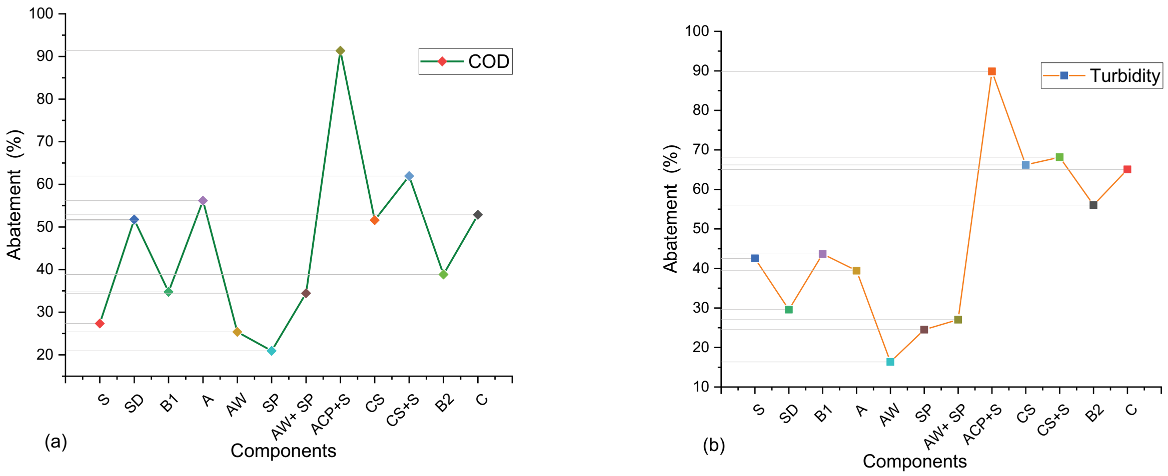

Fig. 10. (a) Chemical oxygen demand abatement and (b) abatement of turbidity.

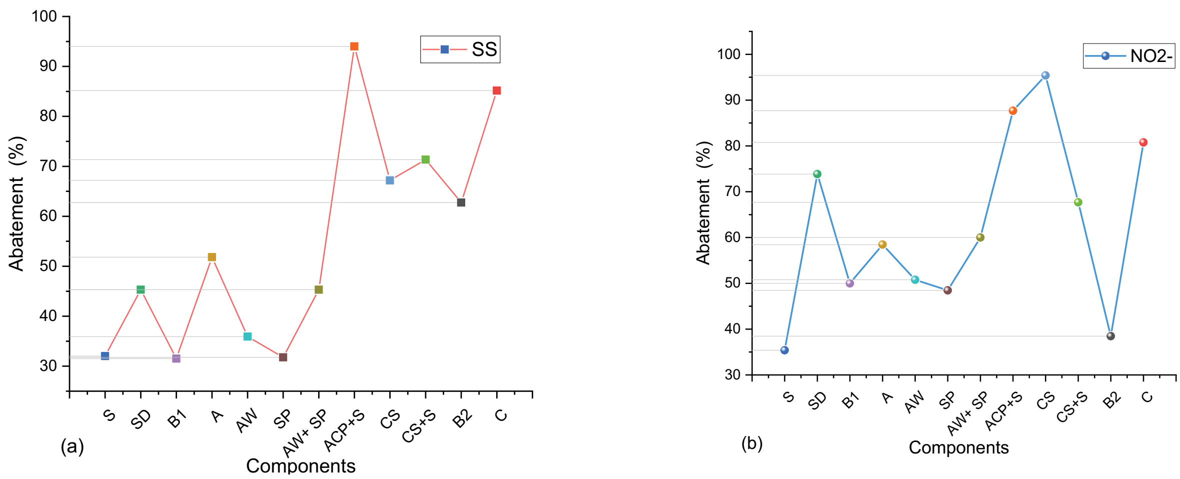

Fig. 11. (a) Abatement of suspended matter and (b) abatement of nitrite.

From the correlation coefficient values, it can be concluded that the adsorption obeys the Langmuir model for both supports.

The identical values of the heterogeneity coefficient n obtained experimentally indicate that the efficiency of clay soil and cactus is similar (n = 0.94 and n = 0.96) (Fig. 15), these values are close to 1 but remain between 0 and 1. These results are in agreement with those obtained previously for the calculation of the % of adsorption, which is 14% for both supports. Moreover, the high values of n confirm the low adsorption rate of clay soil and cactus with respect to our experimental conditions.

4. Conclusions

This research aims to characterize Tunisian leachates and to show that natural adsorbents such as sands, dates or clay soils and certain industrial wastes such as sawdust could be used as a filter medium for leachate treatment. The results obtained show that the studied treated leachates meet the Tunisian indirect discharge standard for the different physico-chemical parameters studied. The filtration through some substrate formed by the clay allows an effective abatement of the physical–chemical pollutant load. In perspective, we propose to proceed to a filtration of leachate on a uniform column, formed essentially by layers of sawdust, clay soils and dates, of well-chosen granulometries, intercalated by natural or industrial adsorbent of normal pH, and this for their efficiency and simplicity of implementation, to better meet the standards of discharge applicable. In this regard, the adsorbents used in this study are still the subject of several more specialized analyzes at the international level. These analyzes in progress, could give us new perspectives. Finally, the development of technologies using organic by-products such as sawdust in the treatment of leachates is an interesting innovation for the future. Indeed, it allows giving a second life to previously discarded waste. On the other hand, the use of organic support still requires studies to optimize the treatments and to test other supports with appreciable purification potential and to drill

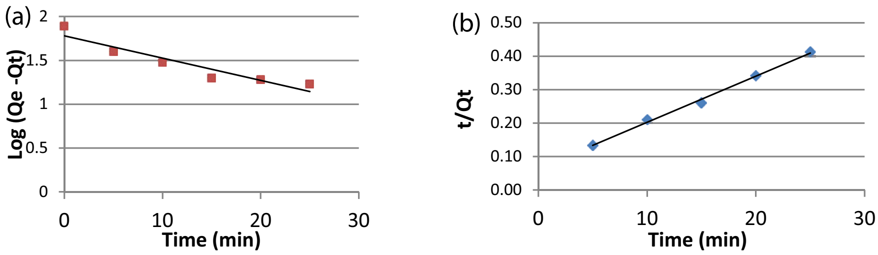

Fig. 12. Pseudo-first-order (a) and pseudo-second-order (b) kinetic model of clay soil.

Table 2
Parameters of kinetic models of adsorption on clay soil

First-order model				Second-order model			
$Q_{e,exp}$ (mg/g)	$Q_{e,cal}$ (mg/g)	K_1 (min^{-1})	R^2	$Q_{e,exp}$ (mg/g)	$Q_{e,cal}$ (mg/g)	K_2 (g mg^{-1} min^{-1})	R^2
76.5	56.21	0.0575	0.892	77.6	76.92	2.64×10^{-3}	0.996

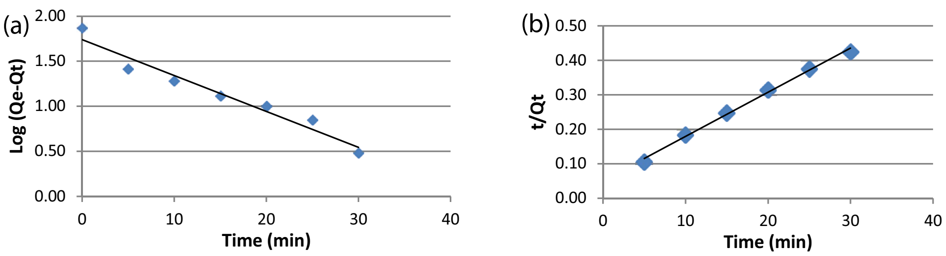

Fig. 13. Pseudo-first-order (a) and pseudo-second-order (b) kinetic model of cactus.

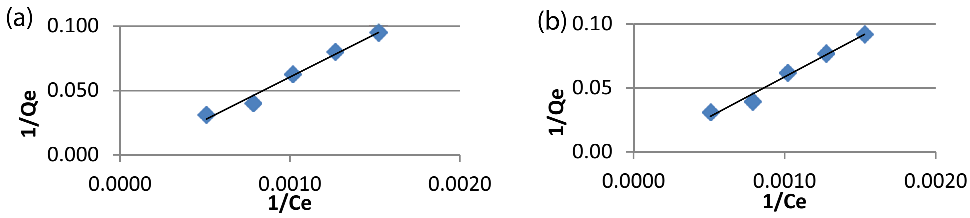

Fig. 14. Langmuir isotherm after adsorption on clay soil (a) and on cactus (b).

Table 3
Parameters of adsorption kinetic models on cactus

First-order model				Second-order model			
$Q_{e,exp}$ (mg/g)	$Q_{e,cal}$ (mg/g)	K_1 (min^{-1})	R^2	$Q_{e,exp}$ (mg/g)	$Q_{e,cal}$ (mg/g)	K_2 (g mg^{-1} min^{-1})	R^2
72.1	55.08	0.01	0.953	73.6	82.30	2.88×10^{-3}	0.995

Table 4
Parameters of adsorption isotherms on clay soil and cactus

Support	Langmuir model			Freundlich model		
	Q_{max} (mg/g)	K_L (L/mg)	R^2	n	K_F (mg/g)	R^2
Clay soil	166	9.1×10^{-5}	0.981	0.94	96.38	0.969
Cactus	250	6.36×10^{-5}	0.980	0.96	79.25	0.966

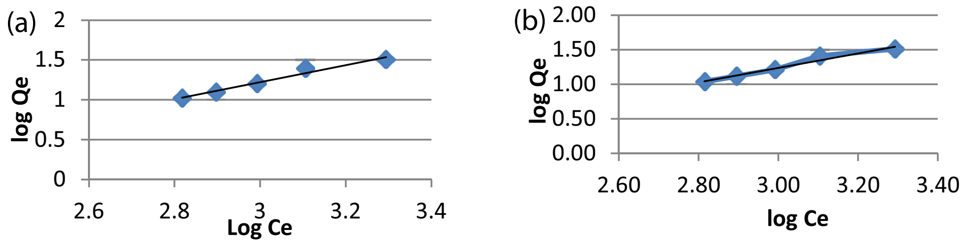

Fig. 15. Freundlich isotherm after adsorption on clay soil (a) and on cactus (b).

configurations and possible combinations between these supports in order to improve their profitability.

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