

Performance of sand and granular activated carbon filtration coupling in tertiary urban wastewater treatment in Algeria

Racha Medjda Bouchenak Khelladi^{a,*}, Abdelghani Chiboub Fellah^a, Maxime Pontié^b, Mehri Shabani^{b,c}, Fatima Zohra Guellil^d

^aLaboratoire de Valorisation des Ressources en eau (V.R.E), Université de Tlemcen, BP 119, 13000 Tlemcen, Algérie, emails: rashamajda@hotmail.fr (R.M. Bouchenak Khelladi), chibabghani@yahoo.fr (A. Chiboub Fellah)

^bFaculté des Sciences, Département de chimie, Université d'Angers, Groupe Analyses & Procédés (GA&P), 2 bd Lavoisier, Angers 49045 cedex 01, France, emails: maxime.pontie@univ-angers.fr (M. Pontié), shabani.mehri@gmail.com (M. Shabani)

^cUniversité Tarbiat Mondares, Tehran Jalal AleAhmad Nasr, P.O. Box: 14115-111, Iran

^dFaculté des sciences, Département de chimie, Université de Tlemcen, BP 119, 13000 Tlemcen, Algérie, email: cfatema@yahoo.fr (F.Z. Guellil)

Received 17 October 2019; Accepted 10 July 2020

ABSTRACT

This work aims to test the efficiency of natural, local and abundant granular media for a tertiary treatment for urban wastewater in Algeria by coupling sand filtration (SF) and granular activated carbon filtration (GACF). The sand media was provided from South Algeria and granular activated carbon (GAC) manufactured by Algerian Company. The pilot consists of two circular columns; the first one filled with sand and the second one with GAC. The SF column was fed by the urban wastewater from the secondary treatment (activated sludge), 5 h a day (5 d week⁻¹) with a constant flow-rate of 30 L h⁻¹, then 5 L of filtered water fed the GACF column. The water was analyzed daily at the inlet and outlet of each filter (SF, GACF). After sixteen weeks of operation, the coupling of sand and activated carbon allowed a reduction of over 75% of the physical-chemical parameters; suspended solids, turbidity, conductivity, biochemical oxygen demand, chemical oxygen demand, hardness, magnesium, calcium, chlorine, nitrates, and phosphates, whereas only 56% has been reached by SF alone and 38% for GACF alone. It can be concluded that coupling sand to activated carbon is efficient to enhance its removal performance and, consequently, improve wastewater quality. Using natural and local materials for treating wastewater is expected to be a promising alternative based on sustainable development.

Keywords: Sand filter; Granular activated carbon filter; Coupling; Urban wastewater; Tertiary treatment

1. Introduction

Different systems can be used for water treatment; some of them are very simple, while others are more complex and use the latest technologies (membrane filtration, chemical oxidation processes, etc). The system should vary according to the quality of water to be treated and the quality of

water that we want to obtain, that is, water for irrigation, but the population does not always have the means to have access to treated water by such systems. That is why we have to find alternatives that are at the same time simple, economical and conform to the regulation according to the sector that it will be supplied for (urban, agricultural, or industrial sector) [1]. The use of sand filters in the treatment

* Corresponding author.

of domestic wastewater has been known for a long time [2]. Sand filters are a natural media that can be used as a solid filter for the treatment of wastewater [3,4], they allow the retention of solids and the fixation of the biomass that can be developed on the granular material, but also, biodegradation of organic, phosphorus and nitrogen pollutants [5,6].

Particulate retention by granular filtration is considered to involve the transport of particles and their attachment to the medium [7]. Transport involves long-range forces or mechanisms that bring a particle near the surface of the collector (support grain) or particles previously retained. The main mechanisms for transporting particles to a single filter are the interception that occurs when the particles that follow the current lines are stopped when they come into contact with the surface of the filter medium, and then the sedimentation which consists of the deposition of the densest particles on the media by gravity, and the Brownian motion (diffusion) which allows the agglomeration of the particles together within the filter media. On the other hand, the fundamental theories of attachment are based on taking into account the Van der Waals forces and the electrochemical double-layer interaction acting on a particle [8,9]. The efficiency of this process depends on several parameters namely, the porosity of the filter, the size and shape of the grains of the filter, as well as the particles to be removed, the characteristics of the fluid (the viscosity and the density) and the characteristics of the flow (flow rate and Reynolds number), these main factors affect hydrodynamics and fixation rates [7,10].

Activated carbon has also been widely used as active support for the treatment of wastewater. These characteristics make it possible to easily introduce structural modifications leading in particular to the improvement of the porosity and the specific surface area [11,12]. Activated carbon is effective because of its porosity so of its large surface contact with the liquid medium and its enormous surface area ($300\text{--}1,500\text{ m}^2\text{ g}^{-1}$), which allows the adsorption of many elements [13,14]. Adsorption is a surface phenomenon by which a substance (solute or adsorbate) present in a solution is extracted from the liquid or gaseous phase and concentrated on the surface of solid material (adsorbent) [15] by adhering to the surface of the filter material [16]. This separation can be done according to two types of mechanisms: physical adsorption (physisorption) and chemical adsorption (chemisorption). Physical adsorption occurs without modification of the material structure and is perfectly reversible. The adsorbed molecules can be easily desorbed by lowering the pressure (in the case of adsorption in the gas phase) or by increasing the temperature. On the contrary, the chemical adsorption is carried out with the formation of valence bonds between the adsorbate molecules and the chemical groups present on the surface of the adsorbent. The binding energy is much stronger than in the case of physisorption, the phenomenon can be irreversible [15,17]. Several studies have shown that the effectiveness of adsorption depends not only on the characteristics of the activated carbon used but also of the pollutant and the solution [18].

This work aims to study the efficiency of natural granular media on the performance of coupling sand and activated carbon filtration as a tertiary treatment on urban wastewater from Ain El Houtz (Tlemcen-Algeria) wastewater

treatment plant. To achieve this work we have used a filtration pilot (TE 400 manufactured by DELTALAB, Germany), in which we have proceeded to a sand filtration (SF) then followed by a filtration on granular activated carbon filtration (GACF) using two distinct columns. To monitor our study, we have taken as reference the physical–chemical parameters (suspended solids (SS), turbidity, electrical conductivity (EC), pH, temperature, nitrate, phosphate, chlorine, magnesium, hardness, biochemical oxygen demand (BCOD), chemical oxygen demand (COD)) that we have analyzed at the inlet and outlet of each filter (SF, GACF) 5 d a week during sixteen weeks. After proceeding to the assessment of the results/analysis, we can say if the proposed tertiary treatment coupling sand and activated carbon can be or not effective to improve the quality of wastewater and if it presents synergies between both steps associated.

2. Material and method

2.1. Pilot description

The pilot that we have used for filtration on sand is the pilot TE 400 manufactured by DELTALAB (Fig. 1) and provided by the Laboratory of Valorization of Water Resources in Algeria, it is composed of a feed tank (1) (150 L); a filtration column (2) in Altuglas (From AKERMA Group located in France) with an internal diameter of 100 mm and a height of 1,000 mm and two brass support and stop grids with a mesh of 0.5 mm; a support frame (3), two manual inlet and outlet flow control taps or filtrate, twelve piezometric multitubes (4) measuring the pressure in the filter column at different heights, a float flow-meter (5) of the column filtrate outlet circuit, a suspension supply pump (6) and a manual valve for dispensing the top of the filter. A second polyethylene filtration column was used for the filtration on activated carbon, with an internal diameter of 40 mm and a height of 1,000 mm, equipped with a filtrate outlet valve.

2.2. Filter media

For our study, two types of filter media were used, namely sand from Southern Algeria and commercial granular activated carbon (GAC) (SARL PROCHIMA MAGHREB, Tlemcen-Algeria), their characteristics were determined at the laboratory and are presented in Table 1.

The particle size analysis gives important information on the filter medium used for this study, it was carried out according to Standard NF EN 933-1 [19,20] which consists of passing a quantity of the sample through a succession of sieves with decreasing opening diameters by applying vibrations.

The grain size distribution curve is used to estimate the effective diameter D_{10} (where 10% of the weight of sand is less than D_{10}) and D_{60} (where 60% of the weight of sand is less than D_{60}), the uniformity of the grain size distribution (the uniformity coefficient) is calculated by the ratio D_{60} and D_{10} [21].

The ideal sand size for a filter is medium to fine with an effective diameter between 0.3 to 1.5 mm (0.5 mm in our study) and a uniformity coefficient less than 4 to have an adequate hydraulic conductivity and to minimize the risk of clogging [22,23].

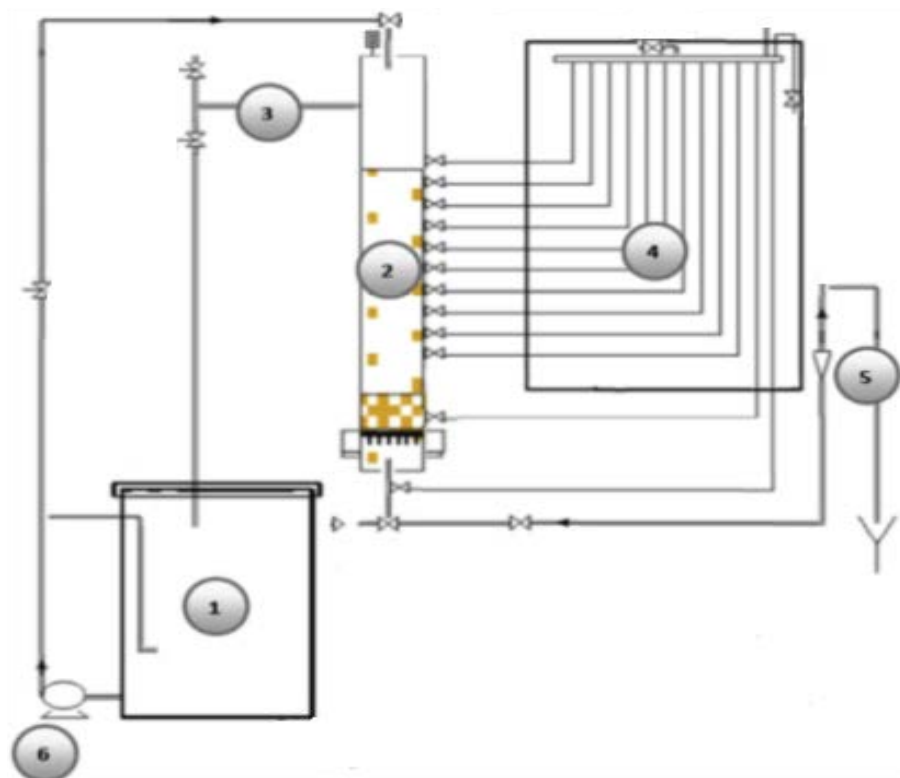


Fig. 1. Pilot TE 400 (manufactured by DELTALAB).

Table 1
Characteristics of the media used

| | Sand | GAC |
|------------------------------|-------|-----|
| D_E (mm) | 0.5 | 1.2 |
| D_{60} (mm) | 1.4 | 1.9 |
| C_{LI} | 2.5 | 1.6 |
| Real density | 2.5 | 1.1 |
| Apparent density | 1.7 | 0.5 |
| EC ($\mu\text{S cm}^{-1}$) | 3,000 | 700 |
| pH | 8.4 | 8 |

The U.S. Environmental Protection Agency recommends that this coefficient should be between 1.3 and 2.5 (2.5 in our study) [20]. Both conditions have been respected in our work.

To avoid the wall effect, the particle size used is chosen so that the ratio between the diameter of the column and the sand grain is greater than 20.7 [24], in our case this ratio is between 70 and 178.

Two other parameters were determined at the laboratory according to Liénard et al. [25] such as the volumic mass (ρ), and the density (d).

2.3. Filtration procedure

The SF pilot is fed with purified wastewater from the Ain El Houtz (34°55' North, 1°19'32" West) wastewater

treatment plant (after secondary treatment by activated sludge and settling) with a filtration rate of 30 L h⁻¹ corresponding to a filtration speed of 3.82 m h⁻¹. The purified wastewater parameters analysis was done at the laboratory presented in Table 2.

For our study, the water first passes through the SF column and then through the activated carbon filtration column by gravity.

Our study took place over sixteen weeks, where the sand filter is fed for 5 h with a flow rate of 30 L h⁻¹ five times a week (we worked 5 d/7, and therefore a daily volume of 150 L which is the useful capacity of the feed tank), to ensure optimal conditions of operation of the column more precisely the regulation of biomass and oxygenation [26]. At the end of each 5 h SF cycle, 5 L of water is collected and then filtered by GAC.

2.4. Sampling and analysis

Samples are taken daily at the inlet and outlet of each filter (SF, GACF) after 5 h cycle and then analyzed at the laboratory.

The main parameters that have been used to evaluate the performance of sand and activated carbon filters are:

- Measurement of the conductivity by a HANNA HI 8633 (Italy) conductivity meter.
- Measurement of turbidity using a HANNA HI 93703 (Italy) turbidimeter.
- Measurement of the temperature using a thermometer.

Table 2
Characteristics of the treated wastewater used (analyzed at the laboratory)

| Parameters | Values |
|------------------------------------------|--------|
| Temperature (°C) | 20–28 |
| Conductivity ($\mu\text{S cm}^{-1}$) | 899 |
| Turbidity (NTU) | 49 |
| COD (mg L^{-1}) | 89 |
| SS (mg L^{-1}) | 100 |
| pH | 6.4 |
| BCOD ₅ (mg L^{-1}) | 63 |
| Calcium (mg L^{-1}) | 15 |
| Magnesium (mg L^{-1}) | 300 |
| Hardness (mg L^{-1}) | 325 |
| Chlorine (mg L^{-1}) | 215 |
| Nitrate (mg L^{-1}) | 13 |
| Phosphate (mg L^{-1}) | 21 |

- Measurement of COD by oxidation with potassium dichromate.
- Measurement of SS with the gravimetry method.
- PH measurement using a WTW pH 3110 pH meter.
- Measurement of BCOD₅ using a BDOCmeter by the manometric method.
- Determination of calcium, magnesium hardness by ethylenediaminetetraacetic acid titrimetry.
- Determination of chlorides by the Mohr method titrimetry.
- Determination of nitrates and phosphates by spectrophotometry.
- Measurement of the head loss in the sand (after a 5 h filtration cycle): by direct reading on piezometric tubes placed at different heights of the filter bed; they are expressed in cm of water with a minimum value of head loss of 30 cm and a maximum value of head loss of –30 cm.

The values that will be presented afterward representing the quality of the filtered water each week, are obtained by averaging the analyzes made every day after a 5 h cycle at the rate of 5 d/7. All the steps followed for our study are illustrated in Fig. 2.

2.5. Filter washing procedure

The head losses and the change in turbidity at the filter outlet are an important indicator of filter performance and which determines the operating time of a filter [27], generally, we are interested in the variation of the turbidity at the filter outlet to proceed for backwash washing with water, and washing is stopped when the input and output water turbidities are identical [28].

In our study we did not wash the filters because we wanted to benefit from the filter clogging and the schmutzdecke layer; the biological removal of nitrate or other organic contaminants occurs in this layer during purification of water. This is well established by the fact that the removal of schmutzdecke affects bacterial reductions [29].

3. Results and discussion

3.1. Evolution of head losses

It has been found that the development of head loss varies linearly with time (Fig. 3). Kellil and Bensafia [28] found that this linearity results in a good deep filtration in the filter bed.

Rolland et al. [30] have observed that the head losses are affected by the accumulation of solids in the filter bed during filtration, we observed that the filter clogging was mainly physical because of the deposition of solids which can be very fast; in just a few weeks.

According to Verma et al. [29] when fouling occurs, the head loss across the filter increases and beyond a certain point, maintaining the flow becomes so difficult that the operation of the filter is interrupted and the washing of the filter must be carried out.

Roussel et al. [31] presented a case study on particle filtration; indicating that the general closure process can be considered as a function of the ratio of the pore size of the particle to the mesh, the solid fraction and the number of grains arriving at each mesh hole during an assay.

The clogging of the media filter can also be caused by the formation of particles smaller than pore size results from the formation of hydrodynamic bridges. During this bridging, the particles clog the pores by forming bridges across the pores inlet (Fig. 4), and in this case, the head loss increased when the hydrodynamic force was strong enough to overcome the colloidal repulsion force of the interparticle surface and particle-pore [32].

The increase in the heat loss during the filtration cycle can be explained by two possibilities, the first is that the upper part of the bed has reached a non-retentive stage, that is to say, that no other collection of particles would have occurred in the upper part of the bed and that other deposits would have occurred mainly in the lower part of the bed. This would increase the head loss across the bed, while the head loss in the upper part of the bed would remain constant, the second possibility is that, even if the particle deposits continue at the top of the bed, the increase of shear stresses in the bed can clean the deposits and transport the fragments to the bottom of the bed. It was also concluded that even if the head loss at the top of the filter bed dominates, there is a fairly significant head loss at the bottom of the filter and there are still particles retained in the upper section of the filter [33].

3.2. Evolution of physical–chemical parameters

EC is the parameter that defines the total salinity of a solution, which is directly proportional to the total concentration of soluble salts [34].

During the first six weeks, there is an increase in EC after filtration on the sand bed from 899 to 1,106 $\mu\text{S cm}^{-1}$, at the end of the 6th week when the EC values of the filtrates from SF results are always higher than the purified wastewater values (Fig. 5).

These values were very high at the beginning of the operating period, due to the high concentration of salts [26], coming from the transformation of organic matter into dissolved salts, from the leaching of sand and GAC minerals or other sources of soluble salts by the effect of common ions

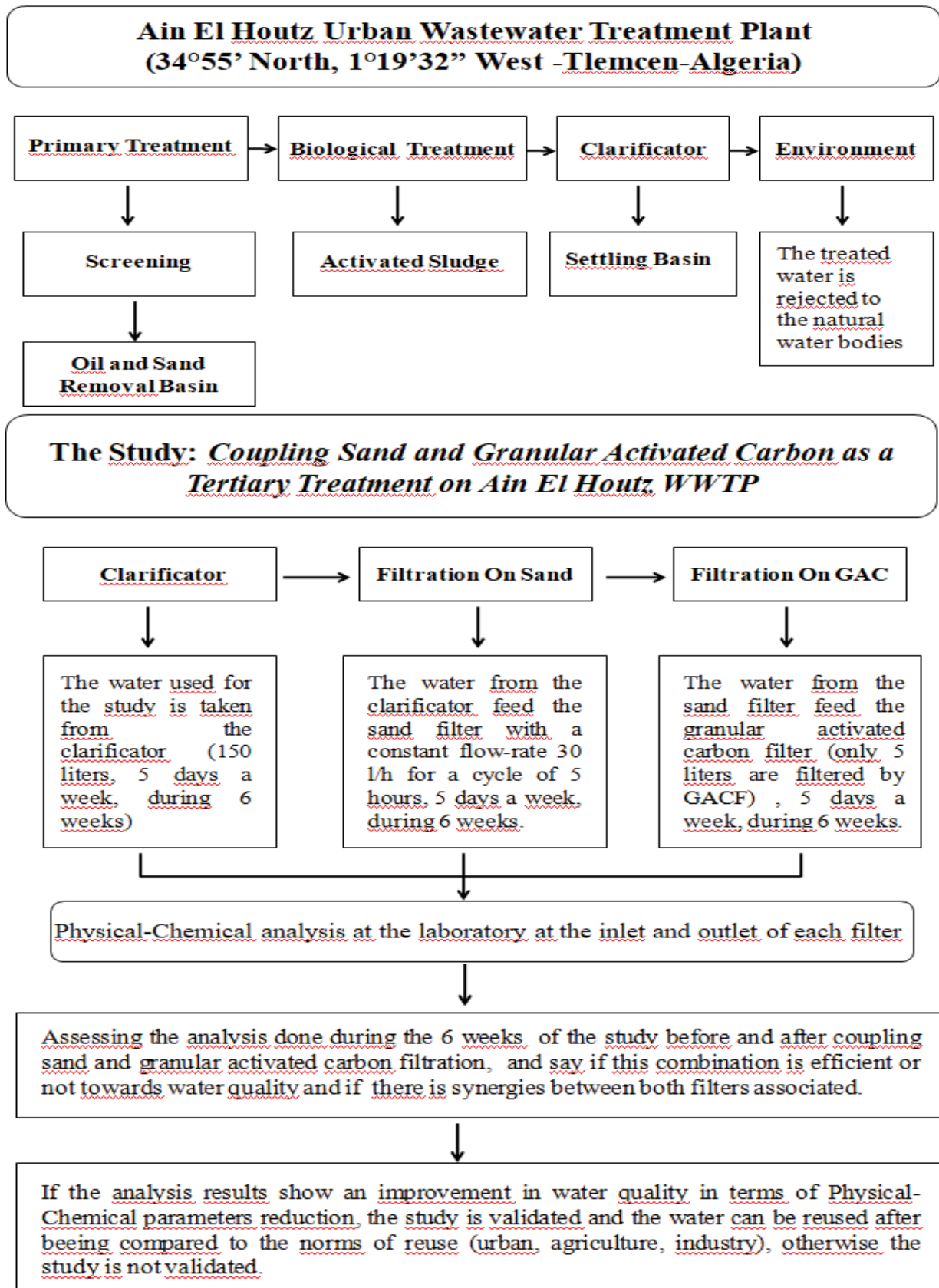


Fig. 2. The diagram on the whole process of the experiment.

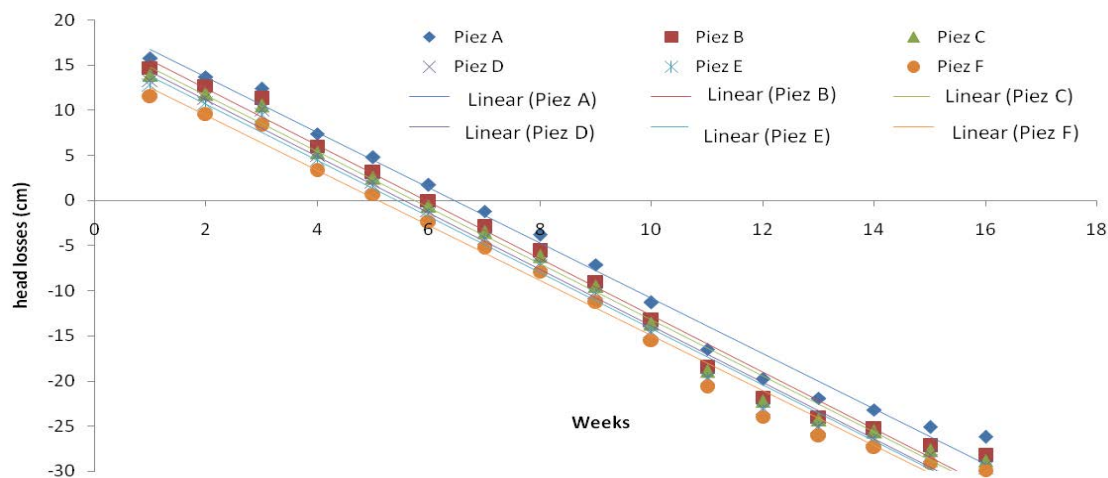


Fig. 3. Head loss evolution during filtration on sand function weeks.

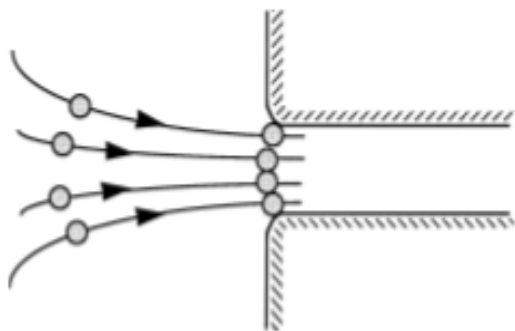


Fig. 4. The particle retention by hydrodynamic bridging [32].

for example (impact related to the composition of the treated effluent) or by microbial activity during the maturation of the filters [35]. This phase represents the leaching stage of the soluble filters, which is the starting point for the leaching of salts existing in the sand and activated carbon [36].

The last ten weeks are characterized by a remarkable decrease in the conductivity values for the sand filter of 54% and this decrease continued after filtration on GAC with 20%. This illustrates the appearance of the clogging phenomenon of the filter by the organic matter as well as the precipitated salts [35,37], these particles retained by the filter will reduce the permeability of the filter medium and consequently the pore space which will facilitate the retention of fine particles (salts) [38].

The pH values of the filtered water are always higher than those of the raw water (purified wastewater), it goes from 6.4 to 8.6 for the sand filter and from 6.4 to 8 for the GAC filter, this increase could be the result of the disappearance of protons, the trapping of organic acids from raw water, and the buffering capacity of sand (8.4) and of GAC (8) and its ability to oppose the variation of pH [37,39], but also, it could be due to the presence of alkaline salts or the dissolution of certain existing salts during the flow of the effluent, which causes certain chemical reactions inside the filters [26,34], as illustrated in Fig. 6. We can also associate the increase of pH by the mineralization of organic

matters by micro-organisms (of the biofilm) formed on the media filter surface (sand and GAC).

Turbidity represents the amount of fine material responsible for the cloudiness of the sample [40,41]. The turbidity of water is a global measure that takes into account all the matters, either colloidal or insoluble, of mineral or organic origin [42].

During filtration on the sand, there is a decrease of the turbidity and SS of 94% and 54% respectively (Figs. 7 and 8), this abatement is due mainly to physical processes (filtration and sedimentation) and sieving in the filtering massive [37]. The consistency of sand performance has improved due to the gradual clogging of smaller pore sizes and increased tension in the top layer of the filter, thus improving the elimination of turbidity by effectively reducing the size of the sand pores of the filter and therefore the retention of more particles [27].

This trend may be consistent with the maturation of the filter, which helps to improve the removal efficiency over time, as the particles previously begin to provide additional removal for suspended particles [43] through the interaction of the contaminants with schmutzdecke (a viscous layer developed on the filter media surface called also biofilm) composed mainly of protozoa, bacteria, algae and other life forms in the filter bed which is considered the main reduction in turbidity [44]. As water passes through the schmutzdecke, suspended particles can be trapped in the filter and the dissolved organic matter is adsorbed and metabolized by the microorganisms [45,46].

It is noted that after 6 weeks of SF, there is a less significant decrease in SS and turbidity, and decreases until the 16th week, this could be explained on the basis that the suspended particles lead to fouling of the pores of the filter which will be closed [47,48], the flow path becomes narrower which significantly increases the shear rate and therefore the part of the materials already deposited on the support will be detached due to increased shear forces resulting from clogging of the filter [49,50], as a result, the retained particles are broken and swept thus the higher the shear rate increases the more difficult it is to accumulate the particles which are more easily detached [29].

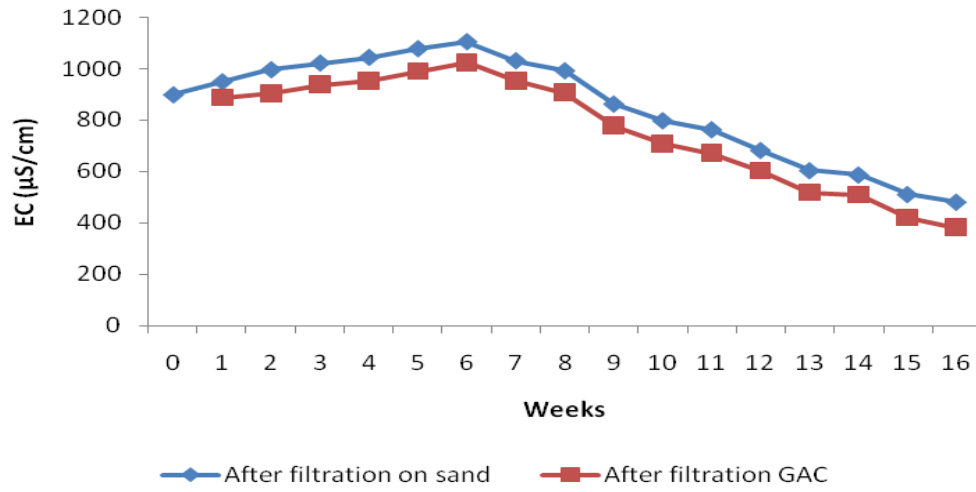


Fig. 5. Evolution of electrical conductivity during sand and GAC filtration according to weeks.

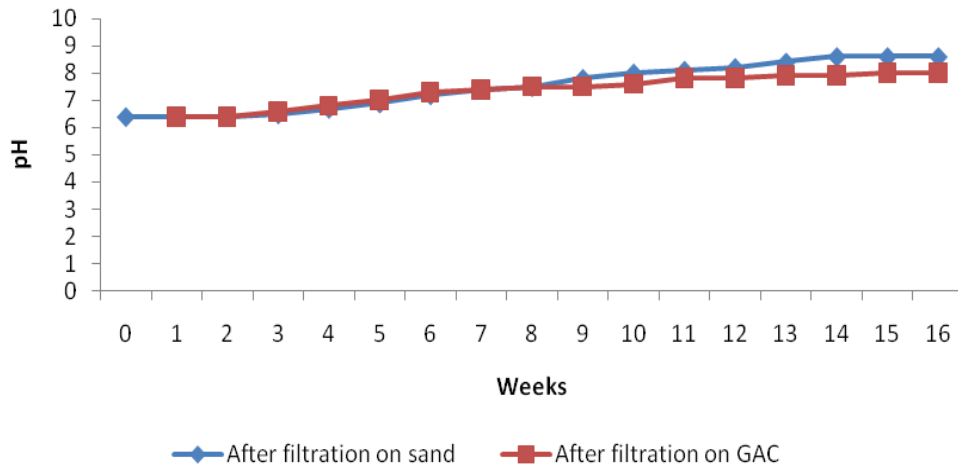


Fig. 6. Evolution of pH during sand and GAC filtration according to weeks.

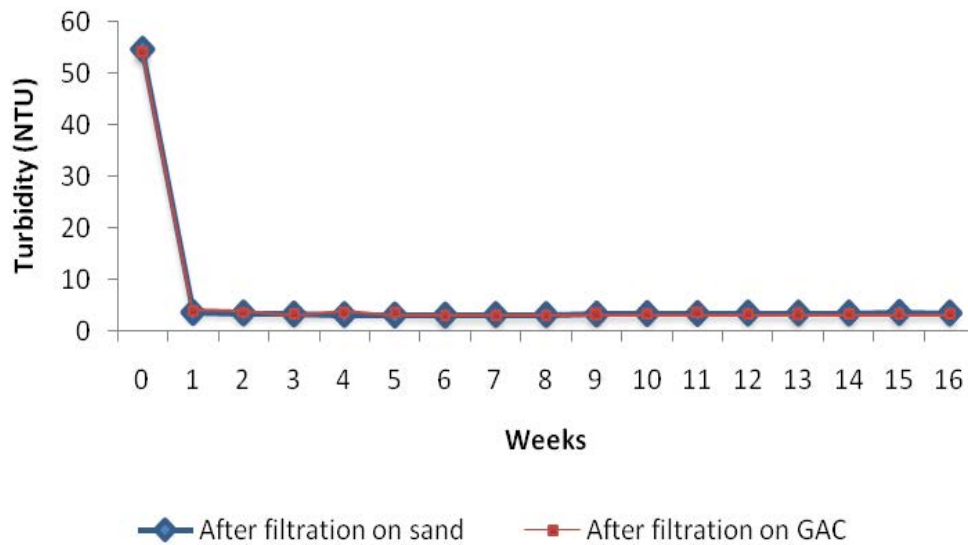


Fig. 7. Evolution of turbidity during sand and GAC function weeks.

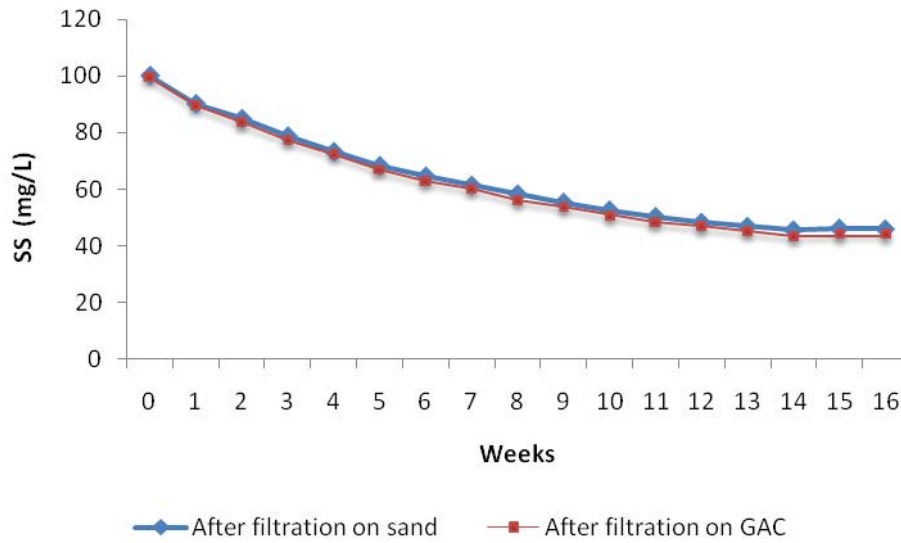


Fig. 8. Evolution of SS during sand and GAC filtration function weeks.

It was also found that the filtration rate which initially was 30 L h⁻¹, decreased linearly and reached 12 L h⁻¹ after 16 weeks of filtration on the sand where only 60 L d⁻¹ was filtered. It can be concluded that after these 6 weeks of filtration, washing is necessary to avoid breaking through the point of the filter and the release of retained particles in the formed cake.

Regarding the results obtained during GACF, it was noted that there is no significant variation in turbidity and SS, because the granulometry of GAC is bigger than sand, so all the particles that have not been retained by sand, cannot be retained by GAC.

The sand filter gives a significant reduction in BCOD and COD of 84% and 73% respectively. After passing through GAC, it was noted that the reduction continued by 51% for BCOD and 38% for COD (Fig. 9).

This phenomenon can be justified by the probable formation of biofilm, in particular at the level of the superficial layer of the two filters, which favors the retention and adsorption of the organic matter and thus its degradation by the bacteria [35,51].

This phase is called the “filter ripening period” where the sand filter begins to crack after 7 d of operation, resulting in a biological activity leading to the degradation of organic matter in the filter. Therefore, the presence of a small amount of organic matter helps to accelerate the maturation of the filters, and thus promote the rapid formation of biomass [34]. This strong reduction in BCOD and COD indicates that good bacterial assimilation takes place in the sand filter and GAC [37], but also that the microbial flora of the biofilm formed has acclimated with the composition of the effluent as weeks go by which facilitates its degradation [52].

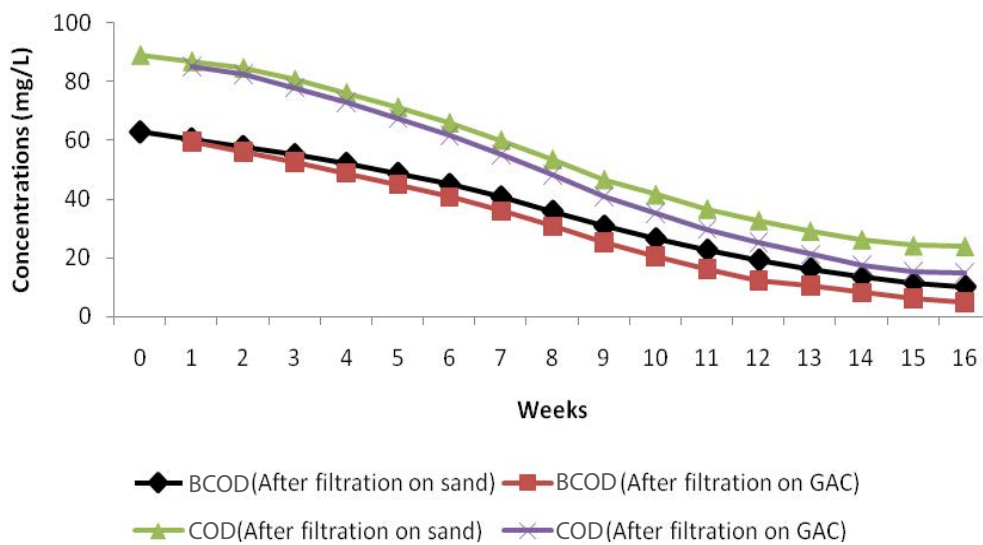


Fig. 9. Evolution of BCOD and COD during sand and GAC filtration according to weeks.

Mechanical entrapment and adsorption of inorganic and organic particles may be the mechanism to reduce BCOD and COD [53], and since the filters have not been washed, it can be considered that a mature filter allows the retention or adsorption to be dominated by the role of the flocs captured in the filters rather than by the clean GAC support [54].

There is a decrease in the improvement of the two filters during the last six weeks, this decrease is accompanied by a 'fatigue' of the filters indicating the beginning of their puncture [55], but the results remain acceptable despite the wear of the filters.

We notice a decrease in hardness of 20%, magnesium 28%, calcium 52%, and chlorine 28% for the sand filter. We also note that the hardness is magnesium because magnesium is significantly higher than calcium.

GAC filtration improved the quality of sand-filtered water by 62% for magnesium, 60% for hardness, 83% for chlorine, and 80% for calcium (Figs. 10 and 11).

The reduction of hardness, magnesium, calcium, and chlorine by the sand filter is mainly due to clogging of the filter which reduces the pore diameter but also by the adsorption phenomenon of these elements on the SS that will be retained by the filter.

On the other hand, the reduction of hardness, magnesium, calcium, and chlorine by the GAC filter is due to its large adsorption surface which gives it the possibility of retention of these elements. The reduction of magnesium and calcium can be done by ion exchange which depends on the overall charge of GAC. The reaction takes place between the functional groups of the surface of the bio-adsorbent (GAC) and the elements of the solution comprising a positive charge (magnesium and calcium) in aqueous solution, through the complexation or the cationic exchange with the negative charges of the surface of the adsorbent and thus it can be expected that GAC promotes the removal of cations such as magnesium and calcium [48,56].

During the last six weeks, a slight reduction with the sand filter, which may be due to the release of the particles previously retained, and the decrease in the observed efficiency of the GAC filter over time can be explained by the saturation of the activated carbon [49].

We note an increase in nitrates and a small reduction of phosphates by 7% in the first five weeks of SF (Fig. 12). This trend is more in water nitrate enrichment than biological denitrification, which means that in this filter, the nitrification reaction that occurs during the six weeks of operation seems to outweigh biological denitrification [57]. Nitrifying microorganisms are known to be the dominant microbial community population on the sand filter [58] and their activity is inhibited by low temperatures as observed by Andersson et al. [59] and Kihn et al. [60] who have shown that the major impact of low water temperature is a decrease in a biological activity which is characterized by a decrease in ammonia removal and not by a complete loss of biomass and that the biomass was still present in the filter even after use in cold water for 5 months, the temperature in our study varies from 20°C to 28°C which explains the good nitrification of water and the nitrate increase in filtered water.

The small reduction of phosphates is due to the aerobic environment in the filter which does not allow the microorganisms to consume the oxygen of the phosphates (PO_4^{3-}) and thus to eliminate them because the elimination of phosphorus requires an anaerobic condition [61].

From the 6th to the 14th week, a strong reduction of nitrates of 83% and phosphates of 67% is noted, beyond the sixth week settles more favorable conditions of denitrification and de-phosphatation, the medium saturated with organic matter becomes more and more anoxic to favor the displacement of the oxygen of the phosphates and the nitrates (breathing of the nitrates) employing denitrifying bacteria [57].

During the last two weeks, there is a slight increase of nitrates and phosphates, these results could be due to the

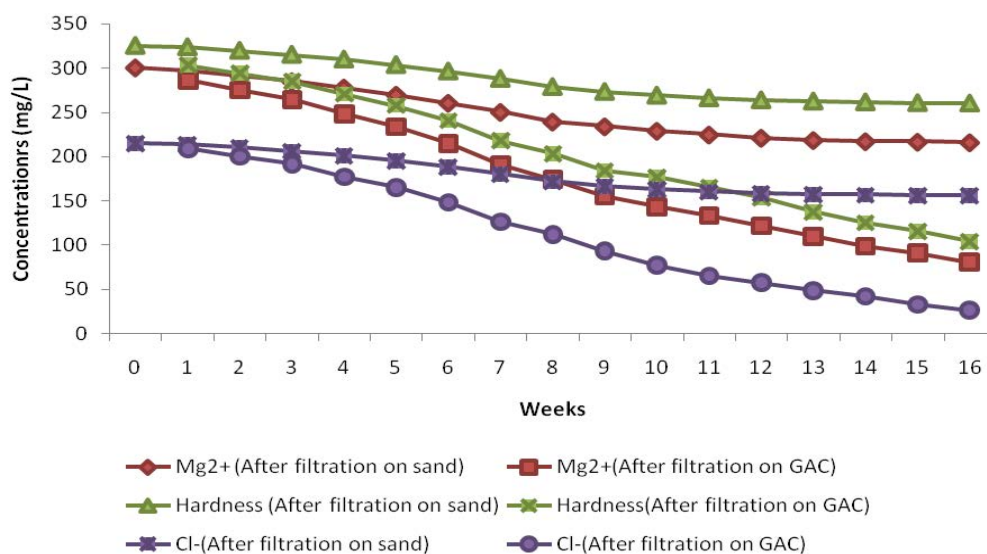


Fig. 10. Evolution of Mg^{2+} , Cl^- and hardness during sand and GAC filtration according to weeks.

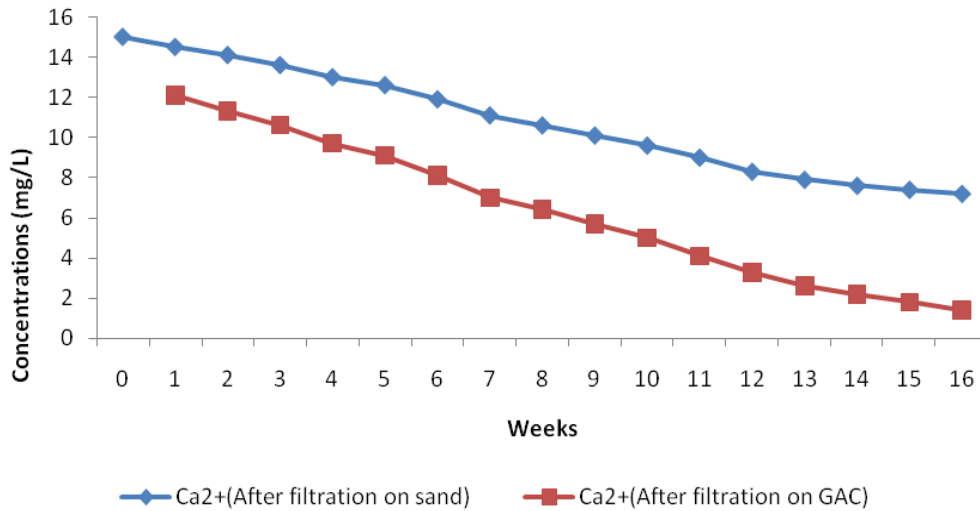


Fig. 11. Calcium evolution during sand and GAC filtration according to weeks.

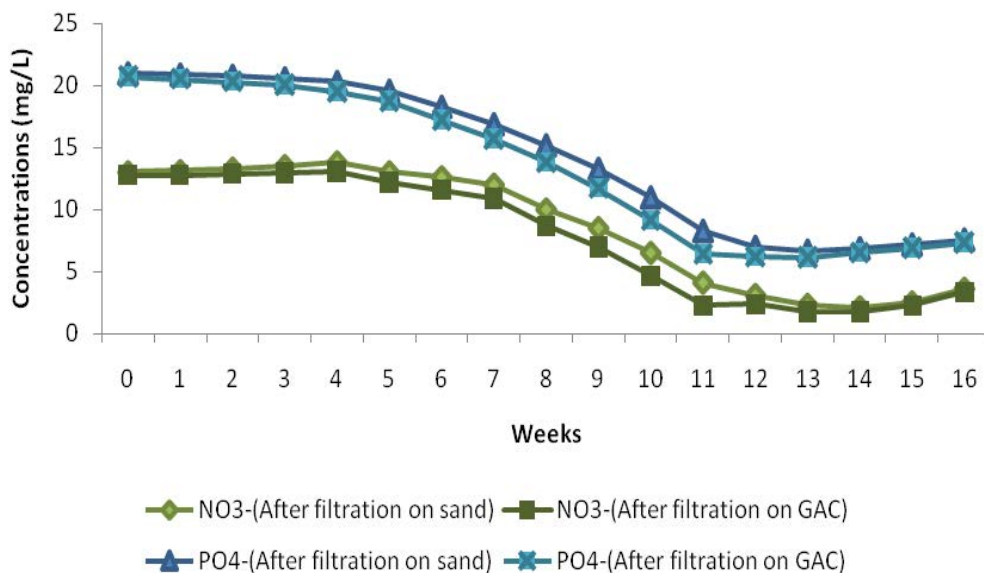


Fig. 12. Evolution of phosphates and nitrates during sand and GAC filtration according to weeks.

saturation of the filtering surface in nitrogen and phosphorus organic matter present in the raw water and in the insufficiency of oxygen, which causes the inhibition of the bacterial activity which can no longer achieve the transformation of these compounds, hence the release of nitrate caused by the massive presence of ammonium and the stabilization of the organic nitrogen contained in the layer of sand [62]. The filter should be given a long operating time to more effectively remove nitrates. Paradoxically, the longer the operating time increases, the more ammonium ions are released [57].

Concerning the GAC filtration of nitrates and phosphates, there is a slight reduction of nitrates of 10% and 7% for phosphates during the first 7 weeks, this reduction could be explained by the fact that when the pH of the water is acidic, the negative charge on the surface of the GAC is

reduced due to the mineralization and the excess in H⁺ protons in solution and makes the number of positive charges increases which will favor the adsorption of the anions (nitrates NO₃⁻ and phosphates PO₄³⁻) due to electrostatic attraction [63]. The pH of the water in our study varies during this interval from 6.4 to 7.4, hence the small reduction of nitrates and phosphates because when the pH tends towards the neutrality the charges on the surface of the GAC are null and therefore the electrostatic effect is canceled.

From the 8th week, a good reduction of nitrates of 43% and phosphates of 22% is noticed. This reduction is not due to the electrostatic effect observed in the previous phase because when the pH increases the charges on the surface of the GAC are negative which do not favor the retention of nitrates and phosphates anions [64], but rather the ammonium cations (NH₄⁺) which will be nitrified during the anoxic

phase in the filter and then retained by the GAC [65], in our study during this interval the pH went from 7.4 to 8.6.

Adsorption is the main mechanism for the removal of phosphorus and nitrates, which is attributed to the large surface area of GAC supporting high biomass density [29], but also by precipitation/fixation reactions [21].

4. Interest of the coupling SAND/GAC

The combination of sand and GAC for wastewater filtration allows interesting reduction yields of physical-chemical parameters. It can be seen (Table 3) that the sand filter alone allows a reduction of more than 50% of the turbidity, COD, SS, BCOD, calcium, nitrates and phosphates, and for the GAC filter allows a reduction of more than 50% of the BCOD, calcium, magnesium, hardness, and chlorine.

It can be said that the sand filter does not react in the same way as the GAC filter towards the reduction of the same parameter. The sand filter gives us an average removal rate of all parameters of 56% and 38% for the GAC, and the coupling of these two allows us to reach 76%. From here we can see the interest of the coupling of filtration on sand and GAC in the tertiary treatment of urban wastewater.

This process is also very interesting for our country Algeria because in addition to its efficiency, it is simple in terms of feasibility, but also it comes as part of the respect for the environment and sustainable development using natural and local materials available in large quantities.

5. Conclusion and perspectives

The tests conducted during this study aimed to check the interest and performance in couplings and GACF on the improvement of the physical-chemical quality of Ain El Houtz wastewater (Tlemcen-Algeria). The first results obtained in the present study are promising because there has been a high improvement in the quality of the water.

The EC was reduced by 47% for the sand filter and this decrease was prolonged after GAC filtration by 21%. For pH, it was increased during SF from 6.4 to 8.6 and 6.4 to 8 for GAC filtration due to the buffering capacity of the two filter media.

Concerning the turbidity and the SS, there was a 94% and 54% decrease respectively after filtration on the sand, on the other hand, no improvement was observed for these two latter after filtration on GAC.

For BCOD and COD, they were reduced by 84% and 73% respectively for the sand filter and this reduction continued after GAC filtration with 51% for BCOD and 38% for COD. Nitrates and phosphates are reduced by 72% and 64% respectively for the sand filter and by 6% and 4% for the GAC filter. Concerning hardness, magnesium, calcium, and chlorine there is a small reduction after SF of 20%, 28%, 52%, and 28% respectively, and a greater reduction of 60%, 63%, 81%, and 83% respectively for the GAC filter.

It can be concluded that the two filters do not react in the same way towards the elimination of the same element, the sand filter is more efficient for the reduction of organic elements whereas the GAC filter is more efficient to reduce mineral elements and from there we can see the importance and interest of their coupling to achieve this association. As a result of this coupling, we reached a reduction of 54% in SS, 94% in turbidity, 74% in nitrates, 65% in phosphates, 58% in conductivity, 92% in BCOD, 83% of the COD, 68% of the hardness, 73% of the magnesium, 91% of the calcium and 88% of the chlorine. We can estimate that the mean yield of this coupling sand/GAC on all the studied parameters about 76%, which is quite consistent.

This study can be used as a reference to define the “Urban Wastewater Treatment Plant of the Future” in Algeria fitting the sustainable development pillars; coupling sand and GACF improve at the same time water quality to reuse it for irrigation to economize conventional water resources; since agriculture is the major water consumer

Table 3
Comparative table of the results of sand and GAC filter and their efficiency rates

| Parameters | Raw water | After SF | After GACF | Elimination yield after SF | Elimination yield after GACF | Elimination yield of the coupling (SF + GACF) |
|------------------------------------------|-----------|----------|------------|----------------------------|------------------------------|-----------------------------------------------|
| EC ($\mu\text{s cm}^{-1}$) | 899 | 480 | 380 | 47% | 21% | 58% |
| Turbidity (NTU) | 54.6 | 3.6 | 3.5 | 93% | 3% | 94% |
| COD (mg L^{-1}) | 89 | 24 | 14.9 | 73% | 38% | 83% |
| SS (mg L^{-1}) | 100 | 46.4 | 44.2 | 54% | 5% | 56% |
| pH | 6.4 | 8.6 | 8 | – | – | – |
| BCOD ₅ (mg L^{-1}) | 63 | 10 | 4.9 | 84% | 51% | 92% |
| Calcium (mg L^{-1}) | 15 | 7.2 | 1.4 | 52% | 81% | 91% |
| Magnesium (mg L^{-1}) | 300 | 216 | 80 | 28% | 63% | 73% |
| Hardness (mg L^{-1}) | 325 | 260 | 104 | 20% | 60% | 68% |
| Chlorine (mg L^{-1}) | 215 | 156 | 26 | 27% | 83% | 88% |
| Nitrates (mg L^{-1}) | 13 | 3.6 | 3.4 | 72% | 6% | 74% |
| Phosphates (mg L^{-1}) | 21 | 7.6 | 7.3 | 64% | 4% | 65% |
| | | | Mean | 56% | 38% | 76% |

SF: sand filter

GACF: activated granular filter

(agriculture 60%, industry 5%) in Algeria and also respect the environment by using no chemicals but only local and natural materials not harmful neither for humans nor for ecosystems. However, further analysis has to be made to confirm the efficiency of sand and GAC coupling on microbiological parameters (pathogenic micro-organisms), even if in literature it is mentioned that filtration on sand removes 50% bacteria and viruses.

In the same perspective of a filtration treatment, another study could be also considered for the treatment of urban wastewater by a membrane process, specifically, nanofiltration/reverse osmosis which will be considered also as tertiary treatment, for a more important minimization of the pollution parameters of treated wastewater and thus to improve its quality, to reuse for the municipality sector for example and if conforms to the norms in the water supply. Other parameters such as organic micropollutants (drug residues) can also be examined to see how they can be reduced by membrane filtration.

References

- [1] F.H. de Souza, B.S. Pizzolatti, J.M. Schöntag, M.L. Sens, Study of slow sand filtration with backwash and the influence of the filter media on the filter recovery and cleaning, *Environ. Technol.*, 37 (2016) 1802–1810.
- [2] A. Liénard, Y. Racault, *Epuration sur supports granulaires: Principes et mise en œuvre*, EUROVITI, Montpellier, 2003, pp. 26–27.
- [3] O.A. Olafadehan, O.W. Jinadu, L. Salami, O.T. Popoola, Treatment of brewery wastewater effluent using activated carbon, *Int. J. Appl. Sci. Technol.*, 2 (2012) 165–178.
- [4] S.A. Al-Jlil, COD and BOD reduction of domestic wastewater using activated sludge, sand filters and activated carbon in Saudi Arabia, *Biotechnology*, 8 (2009) 473–477.
- [5] J.M. Hua, P.L. An, J. Winter, C. Gallert, Elimination of COD, microorganisms and pharmaceuticals from sewage by trickling through sandy soil below leaking sewers, *Water Res.*, 37 (2003) 4395–4404.
- [6] M.L. Weber-Shirk, Enhancing slow sand filter performance with an acid-soluble seston extract, *Water Res.*, 36 (2002) 4753–4756.
- [7] N.-E. Sabiri, E. Monnier, V. Raimbault, A. Massé, V. Séchet, P. Jaouen, Effect of filtration rate on coal-sand dual-media filter performances for microalgae removal, *Environ. Technol.*, 38 (2017) 345–352.
- [8] J.K. Kim, J.A. Nason, D.F. Lawler, Influence of surface charge distributions and particle size distributions on particle attachment in granular media filtration, *Environ. Sci. Technol.*, 42 (2008) 2557–2562.
- [9] T. Asano, F.L. Burton, H.L. Leverenz, R. Tsuchihashi, G. Tchobanoglous, *Water Reuse: Issues, Technologies, and Applications*, Metcalf & Eddy, Inc., New York, 2007.
- [10] B. Benmezroua, *Etude Numérique et Expérimentale, à L'échelle Microstructurelle, du Transport Granulaire Dans les Matériaux Poreux Saturés*, INSA de Rennes, France, 2011.
- [11] F. Rogalla, P. Ravarini, G. de Larminat, J. Couttelle, Large-scale biological nitrate and ammonia removal, *Water Environ. J.*, 4 (1990) 319–328.
- [12] A. Bhatnagar, A.K. Minocha, Conventional and non-conventional adsorbents for removal of pollutants from water – a review, *Indian J. Chem. Technol.*, 13 (2006) 203–217.
- [13] L. Dauphin, Développement d'un test rapide pour prédire la performance d'un réacteur à haute concentration de charbon actif recirculé, *École Polytechnique de Montréal*, Canada, 2017.
- [14] R. Singhon, Adsorption of Cu(II) and Ni(II) Ions on Functionalized Colloidal Silica Particles Model Studies for Wastewater Treatment, Thesis for the Degree of Doctor of Science (Chemistry), Franche-Comté University, France, 2014.
- [15] T.F. de Oliveira Penalver, Etude d'un procédé de dépollution basé sur le couplage ozone/charbon actif pour l'élimination des phtalates en phase aqueuse, University of Orléans, Orléans-France, 2011.
- [16] H.P. Shivaraju, H. Egumbo, P. Madhusudan, K.M. Anil Kumar, G. Midhun, Preparation of affordable and multifunctional clay-based ceramic filter matrix for treatment of drinking water, *Environ. Technol.*, 40 (2019) 1633–1643.
- [17] L.-M. Sun, F. Meunier, N. Brodu, M.-H. Manero, *Adsorption: Aspects Théoriques, Techniques de l'ingénieur*, France, 2003.
- [18] A. Dąbrowski, P. Podkościelny, Z. Hubicki, M. Barczak, Adsorption of phenolic compounds by activated carbon—a critical review, *Chemosphere*, 58 (2005) 1049–1070.
- [19] C. Wang, Etude comparative des matériaux de garnissage dans les réacteurs de filtration pour l'assainissement non collectif, Thesis for the Degree of Doctor of Water, Soil and Environment, Limoges University, France, 2015.
- [20] USEPA, APHA, AWWA, WEF, Standard Methods for the Examination of Water and Wastewater, United States Environmental Protection Agency, American Public Health Association, American Water Works Association, Water Environment Federation, Washington, DC, 1998, pp. 3–37.
- [21] C.A. Arias, M. Del Bubba, H. Brix, Phosphorus removal by sands for use as media in subsurface flow constructed reed beds, *Water Res.*, 35 (2001) 1159–1168.
- [22] F. Chen, J. Tao, K. Mancl, Sand Size Analysis for Onsite Wastewater Treatment System, Determination of Sand Effective Size and Uniformity Coefficient, The Ohio State University, USA, 2008. Available at: <https://ohioline.osu.edu/factsheet/aex-757>
- [23] M. Achak, L. Mandi, N. Ouazzani, Removal of organic pollutants and nutrients from olive mill wastewater by a sand filter, *J. Environ. Manage.*, 90 (2009) 2771–2779.
- [24] M.C. Moran, D.C. Moran, R.S. Cushing, D.F. Lawler, Particle behavior in deep-bed filtration: Part 2—particle detachment, *J. Am. Water Works Assn.*, 85 (1993) 82–93.
- [25] A. Liénard, H. Guellaf, C. Boutin, Choice of the sand for sand filters used for secondary treatment of wastewater, *Water Sci. Technol.*, 44 (2001) 189–196.
- [26] G. Yamina, A. Abdeltif, T. Youcef, H.M. Mahfoud, G. Fatiha, B. Lotfi, A comparative study of the addition effect of activated carbon obtained from date stones on the biological filtration efficiency using sand dune bed, *Energy Procedia*, 36 (2013) 1175–1183.
- [27] P.D. Davies, A.D. Wheatley, Pilot plant study of alternative filter media for rapid gravity filtration, *Water Sci. Technol.*, 66 (2012) 2779–2784.
- [28] A. Kellil, D. Bensafia, Removal of phosphates by direct filtration on sand bed, *J. Water Sci.*, 16 (2003) 317–332.
- [29] S. Verma, A. Daverey, A. Sharma, Slow sand filtration for water and wastewater treatment – a review, *Environ. Technol. Rev.*, 6 (2017) 47–58.
- [30] L. Rolland, P. Molle, A. Liénard, F. Bouteldja, A. Grasmick, Influence of the physical and mechanical characteristics of sands on the hydraulic and biological behaviors of sand filters, *Desalination*, 248 (2009) 998–1007.
- [31] N. Roussel, T.L.H. Nguyen, P. Coussot, General probabilistic approach to the filtration process, *Phys. Rev. Lett.*, 98 (2007) 114502.
- [32] V. Ramachandran, H.S. Fogler, Plugging by hydrodynamic bridging during flow of stable colloidal particles within cylindrical pores, *J. Fluid Mech.*, 385 (1999) 129–156.
- [33] Z. Skaf, O.F. Eker, I.K. Jennions, A simple state-based prognostic model for filter clogging, *Procedia CIRP*, 38 (2015) 177–182.
- [34] F. Gherairi, B. Hamdi-Aissa, Y. Touil, M. Hadj-Mahammed, H. Messrouk, A. Amrane, Comparative study between two granular materials and their influence on the effectiveness of biological filtration, *Energy Procedia*, 74 (2015) 799–806.
- [35] K. Khengaoui, M.H. Mahammed, Y. Touil, A. Amrane, Influence of secondary salinity wastewater on the efficiency of biological treatment of sand filter, *Energy Procedia*, 74 (2015) 398–403.
- [36] M.G. Healy, M. Rodgers, P. Burke, Quantification of biofilm build-up in filters when intermittently loaded with

- low-strength synthetic wastewater, *Desalination*, 271 (2011) 105–110.
- [37] M. Achak, N. Ouazzani, L. Mandi, Élimination des polluants organiques des effluents de l'industrie oléicole par combinaison d'un filtre à sable et un lit planté, *Rev. Des Sci. l'eau/J. Water Sci.*, 24 (2011) 35–51.
- [38] E.J. Roth, B. Gilbert, D.C. Mays, Colloid deposit morphology and clogging in porous media: fundamental insights through investigation of deposit fractal dimension, *Environ. Sci. Technol.*, 49 (2015) 12263–12270.
- [39] M. Ben Abbou, M. El Haji, Treatments by electrocoagulation-filtration of uncontrolled leachate discharge from the city of Taza and re-use in the germination of *sorghum* and *alfalfa*, *Int. J. Innovation Appl. Stud.*, 9 (2014) 355–366.
- [40] B. Lipták, *Instrument Engineers' Handbook: Process Measurement and Analysis*, CRC Press, Boca Raton, Florida, 2003.
- [41] C. Baudequin, Design of a Mobile Post-Treatment Unit for the Water used During Fire Extinguishment, *Chemical and Process Engineering*, Ecole Centrale Paris, 2011.
- [42] B. Thayer, K. Riahi, H. Boudhraa, Élimination de la turbidité par oxygénation et filtration successives des eaux de la station de Sfax (Sud de la Tunisie), *Rev. Des Sci. l'eau/J. Water Sci.*, 20 (2007) 355–365.
- [43] J.L. Darby, D.F. Lawler, T.P. Wilshusen, Depth filtration of wastewater: particle size and ripening, *Res. J. Water Pollut. Control Fed.*, 63 (1991) 228–238.
- [44] C. Li, Y.F. Wu, L.B. Zhang, W. Liu, Treatment Efficiencies of Slow Sand Filtration for Landscape Water, 2010 4th International Conference on Bioinformatics and Biomedical Engineering, IEEE, Chengdu, China, 2010, pp. 1–3.
- [45] CAWST, *BioSand Filter Manual—Design, Construction, Installation, Operation and Maintenance*, Centre for Affordable Water and Sanitation Technology, Canada, 2009.
- [46] L.L. Wu, X. Zhao, Z. Meng, Removal of dissolved organic matter in municipal effluent with ozonation, slow sand filtration and nanofiltration as high quality pre-treatment option for artificial groundwater recharge, *Chemosphere*, 83 (2011) 693–699.
- [47] M.W. Jenkins, S.K. Tiwari, J.D. Darby, D. Nyakash, W. Saenyi, K. Langenbach, The Biosand Filter for Improved Drinking Water Quality in High Risk Communities in the Njoro Watershed, Kenya, Research Brief, Global Livestock CRSP, University of California-Davis, Davis, CA, 2009. Available at: <http://scholar.google.com/scholar?hl=en&btnG=Search&q=intitle:The+BioSand+Filter+for+Improved+Drinking+Water+Quality+in+high+Risk+Communities+in+the+Njoro+Watershed#0> (accessed June 21, 2020)
- [48] T.O. Mahlangu, L. Mpenyana-Monyatsi, M.N.B. Momba, B.B. Mamba, A simplified cost-effective biosand filter (BSFZ) for removal of chemical contaminants from water, *J. Chem. Eng. Mater. Sci.*, 2 (2011) 156–167.
- [49] P. Servais, G. Billen, P. Bouillot, Biological activity in a granular activated carbon filter, *Rev. Des Sci. l'Eau.*, 4 (1991) 483–498.
- [50] B. Tansel, F. Vilar, Enhancement of media filter performance with coagulant use for treatment of diesel oil contaminated surface water, *Desalination*, 173 (2005) 69–76.
- [51] R. Khalaphallah, Greywater Treatment for Reuse by Slow Sand Filtration: Study of Pathogenic Microorganisms and Phage Survival, *Chem. Process Eng.*, Ecole des Mines de Nantes, 2012. Available at: <https://tel.archives-ouvertes.fr/tel-00735857> (accessed June 21, 2020)
- [52] S.M. Riley, D.C. Ahoor, T.Y. Cath, Enhanced biofiltration of O&G produced water comparing granular activated carbon and nutrients, *Sci. Total Environ.*, 640–641 (2018) 419–428.
- [53] T.B. Bagundol, A.L. Awa, M.R.C. Enguito, Efficiency of slow sand filter in purifying well water, *J. Multidiscip. Stud.*, 2 (2013), <https://doi.org/10.7828/jmds.v2i1.402>.
- [54] G.J. Williams, B. Sheikh, R.B. Holden, T.J. Kouretas, K.L. Nelson, The impact of increased loading rate on granular media, rapid depth filtration of wastewater, *Water Res.*, 41 (2007) 4535–4545.
- [55] N. Djedidi, A. Hassen, Propriétés physiques des sols et pouvoir colmatant des eaux usées en fonction de leur degré de traitement, *Cah. - ORSTOM, Ser. Pedol.*, 26 (1991) 3–10. Available at: http://horizon.documentation.ird.fr/exl-doc/pleins_textes/cahiers/PTP/35396.PDF (accessed June 21, 2020)
- [56] K.D. Belaid, S. Kacha, Étude cinétique et thermodynamique de l'adsorption d'un colorant basique sur la sciure de bois, *Rev. Des Sci. l'Eau.*, 24 (2011) 131–144.
- [57] O.B. Yapo, V. Mambo, E.J.-C. Meledje Djedjess, M.J. Ohou, A. Seka, A.S. Tidou, P.V. Houenou, Searching for parameters optimising the biological denitrification of nitrate-and ammonium-rich well waters by private slow sand filtration reactors, *Eur. J. Sci. Res.*, 26 (2009) 565–576. Available at: <http://www.eurojournals.com/ejsr.htm> (accessed June 21, 2020)
- [58] K.S. Nitzsche, P. Weigold, T. Lösekann-Behrens, A. Kappler, S. Behrens, Microbial community composition of a household sand filter used for arsenic, iron, and manganese removal from groundwater in Vietnam, *Chemosphere*, 138 (2015) 47–59.
- [59] A. Andersson, P. Laurent, A. Kihn, M. Prévost, P. Servais, Impact of temperature on nitrification in biological activated carbon (BAC) filters used for drinking water treatment, *Water Res.*, 35 (2001) 2923–2934.
- [60] A. Kihn, A. Andersson, P. Laurent, P. Servais, M. Prévost, Impact of filtration material on nitrification in biological filters used in drinking water production, *J. Water Supply Res. Technol. AQUA*, 51 (2002) 35–46.
- [61] A. Torrens, P. Molle, C. Boutin, M. Salgot, Impact of design and operation variables on the performance of vertical-flow constructed wetlands and intermittent sand filters treating pond effluent, *Water Res.*, 43 (2009) 1851–1858.
- [62] S.R. Smith, V. Woods, T.D. Evans, Nitrate dynamics in biosolids-treated soils. II. Thermal-time models of the different nitrogen pools, *Bioresour. Technol.*, 66 (1998) 151–160.
- [63] D.-W. Cho, C.-M. Chon, Y.J. Kim, B.-H. Jeon, F.W. Schwartz, E.-S. Lee, H. Song, Adsorption of nitrate and Cr(VI) by cationic polymer-modified granular activated carbon, *Chem. Eng. J.*, 175 (2011) 298–305.
- [64] O. Khelil, A.E.K. Ouakouak, L. Youcef, S. Achour, Denitrification Des Eaux Souterraines Par Adsorption Sur Charbon Actif Et Par Coagulation-Floculation Au Sulfate D'Aluminium, *LARHYSS J.*, (2015) 181–190.
- [65] I. Bruch, J. Fritsche, D. Bänninger, U. Alewell, M. Sendelov, H. Hürlimann, R. Hasselbach, C. Alewell, Improving the treatment efficiency of constructed wetlands with zeolite-containing filter sands, *Bioresour. Technol.*, 102 (2011) 937–941.