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NITROGEN CONVERSION AND REMOVAL BY BIOLOGICAL AERATED FILTRATION

- Summary of Doctoral Thesis -

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ABSTRACT

NITROGEN CONVERSION AND REMOVAL BY BIOLOGICAL AERATED FILTRATION

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The general goal of current research was to explore the performance of nitrogen conversion and removal by biological aerated filtration. The laboratory experiments by three identical pilot-scales downflow of biological aerated filter system (BAFs) using three different filtration media types, 0.78 ± 0.60 mm activated carbon-based material bed, 0.95 ± 0.58 mm sand-based material bed, and 3.28 ± 2.14 mm ceramic particle-based material bed as biomass support as attached growth zones in treating of municipal wastewater to the developing local materials bed, low cost, for improving the effluent quality before discharge to the aquatic environment.

Throughout the duration of the experiments, all the three pilot-scales downflow BAFs were continuously operated fed with raw municipal wastewater containing various pollutants, e.g. nitrate-N, nitrite-N, and ammonium-N. All the three pilot-scales downflow BAFs were operated at ambient air temperature ranged from 8 to 29 °C with a mean of 17.93±7.27 °C. The mean influent flowrates were 0.18 L/min, and air: liquid ratio of 10:1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and N loading rates of the three pilot-scales downflow BAFs was $0.0022\pm0.00~kg~NH_4^+-N/m^3.d,~0.0072\pm0.00~kg~TKN/m^3.d,~0.0434\pm0.01kg~tCOD/m^3.d,~0.0285\pm0.00~kg~sCOD/m^3.d,~and~0.0931\pm0.17~kg~SS/m^3.d.$

Assessed the pilot-scale downflow BAFs performance using activated carbon-based filtration media bed for nitrogen conversion and removal. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 90.11%, 90.10%, 91.84%, 86.37%, 76.26%, respectively for NO₃-N, NO₂-N, NH₄+N, TKN, and TN. Furthermore, was able to nitrify 0.24 \pm 0.21 kg NH₄+N/m³.d, 0.19 \pm 0.18 kg NO₂-N/m³.d, 0.15 \pm 0.15 kg NO₃-N/m³.d, 0.78 \pm 0.54 kg TKN/m³.d, and 2.14 \pm 2.35 kg TN/m³.d. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in

terms of tCOD, sCOD, and SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 91.89%, 91.47%, 94.24%, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.07 ± 2.42 kg tCOD/m³.d, 3.31 ± 1.06 kg sCOD/m³.d, and 5.72 ± 4.08 kg SS/m³.d.

Appraised the pilot-scale downflow BAFs performance of using sand-based filtration media bed for nitrogen conversion and removal. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 87.74%, 89.84%, 90.28%, 84.82%, and 75.29%, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN, TN. Furthermore, was able to nitrify 0.24±0.22 kg NH₄⁺-N/m³.d, 0.19±0.18 kg NO₂⁻-N/m³.d, 0.15±0.15 kg NO₃⁻-N/m³.d, 0.77±0.54 kg TKN/m³.d, and 2.11±2.35 kg TN/m³.d. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 91.60%, 90.96%, 93.34%, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.06±2.42 kg tCOD/m³.d, 3.28±1.08 kg sCOD/m³.d, and 5.66±4.03 kg SS/m³.d.

Evaluated the performance of pilot-scale downflow BAFs using ceramic particle-based filtration media bed for nitrogen conversion and removal. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 85.17%, 88.32%, 89.03%, 83.69%, and 74.82%, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N,TKN, TN. Furthermore, was able to nitrify 0.23 ± 0.22 kg NH₄⁺-N/m³.d, 0.18 ± 0.18 kg NO₂⁻-N/m³.d, 0.15 ± 0.15 kg NO₃⁻-N/m³.d, 0.76 ± 0.54 kg TKN/m³.d, and 0.15 ± 0.15 kg TN/m³.d. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 90.51%, 90.51%, 83.27%, respectively for tCOD, sCOD, SS. And the mean mass removal was 0.02 ± 0.02 kg sCOD/m³.d, and 0.02 ± 0.02 kg sCOD/m³.d

Results of current study revealed that all three-filtration media used in the pilot-scales downflow BAFs with three different sCOD/TKN ratios (2.8, 4.0, 4.5) indicate that as the C/N ratio increased, the N (NO_3^--N , NO_2^--N , and NH_4^+-N) elimination (%) decreased linearly, as well as, increased the C/N ratio, the mass of the N removed (NH_4^+-N , NO_2^--N , and NO_3^--N) per unit volume (NH_4^+-N , NO_2^--N , and NO_3^--N) levels increased as the C/N ratio increased.

The experimental results with three different sCOD/TKN ratios (2.8, 4.0, 4.5) indicate that as the C/N ratio increased, the COD (sCOD, and tCOD) loading decreased, and COD (sCOD, and tCOD) removal (%) decreased, Correspondingly, the C/N ratio decreased, the mass of the COD removed (sCOD, and tCOD) per unit volume (kg/m³.d), and removal (%) increased.

The results of current research revealed that all three-filtration media used in the pilot-scales downflow BAFs was extremely well in the nitrogen conversion and removal. However, of the three-filtration media-based bed types, the pilot-scale downflow BAFs which used a 0.78±0.60 mm activated carbon-based filtration media bed recorded the highest percent of nitrogen elimination efficacy and mass.

Keyword: Biological aerated filtration; Nitrogen; Nitrates; Nitrites; Ammonia.

1 Background and Objectives

1.1 General

This chapter presented the background of this study, including the nitrogen formula and problems the nitrogen discharged from point sources and non-point sources. Also, this chapter presented the wastewater technologies for nitrogen removal, including a description of the BAFs. Finally, this chapter presented a problem statement of the research, the scope of the research, this allows defining the key objective of current thesis, in addition to various working goals consequent from this key objective. The structure of the thesis presented at the end of the chapter.

1.2 Background of the Thesis

1.2.1 Nitrogen Formula

Nitrogen occurs in the environment in various forms. The nitrogen cycle controls the conversion and transport of these compounds in the biosphere (Figure 1.1) (Delwiche, 1970) [50]. Nitrogen can form different compounds as it shows various oxidation states. The majority of oxidation state transitions in the environment caused by biological processes.

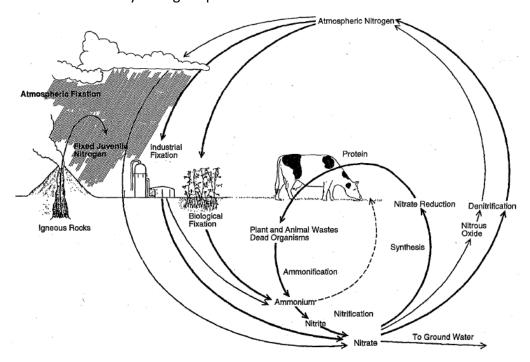


Figure 1.1. The N cycle (U.S. Environmental Protection Agency, 1993) [53], [202].

N compound	Formula
Ammonia	NH_3
Ammonium ion	$\mathrm{NH_4}^+$
Nitrite Ion	NO_2^-
Nitrate ion	NO ₃ -
Nitrogen gas	N_2

Nitrogen fixation ensures that inert, gaseous nitrogen is involved in the chemical compound to be used by animals or plants. Specialized microorganisms and associations among microorganisms perform biological fixation of N_2 gas to organic nitrogen (McCarty & al., 1967) [121]. A smaller role has lighting and industrial N fixation.

Ammonification process is the conversion from organic N to ammonium. A significant hydrolysis procedure includes urea, a urine N-compound:

$$H_2NCONH_2 + 2H_2O \xrightarrow{\qquad \qquad} (NH_4)_2CO_3$$
urease
urea ammonium carbonate

In general, ammonification develops during the decomposition of animal and plant tissues, as well as animal faeces:

Organic N + Microorganism
$$\rightarrow NH_3/NH_4^+$$
 (Amino acids, protein, etc.)

Synthesis, a biochemical reaction involves the formation of plant protein or other nitrogenous compounds by using ammonium or nitrate compounds:

$$NO_3^- + CO_2 + \text{green plants} + \text{sunlight.}$$
 \longrightarrow protein $NH_3/NH_4^+ + CO_2 + \text{green-plants} + \text{sunlight}$ \longrightarrow protein

Animals aren't capable of nitrogen fixation with certain exceptions, they need protein either from plants or other animals. The biological oxidation of ammonia is called *Nitrification*, which occurs in two stages, first forming nitrite then nitrate. Two particular chemoautotrophic bacteria are involved that utilize inorganic carbon as their carbon source:

Nitrosomonas Nitrobacter

$$NH_4^+ + O_2 \longrightarrow NO_2^- + O_2 \longrightarrow NO_3^-$$

bacteria bacteria

Nitrite Nitrate

These transformations are commonly and rapidly progress to nitrate; the nitrite levels is relatively lower at a time. The produced nitrate might be used in production to enhance plants' growth or decreased by denitrification (Figure 1.1).

Denitrification procedure is biological reducing nitrate to nitrogen gas. The crucial production of nitrogen gas can be formed by several steps of the biochemical pathway. Various heterotrophic bacteria are involved in denitrification, containing an organic C as a source of energy:

$$NO_3^- + Organic C \longrightarrow NO_2^- + organic C \longrightarrow N_2 + CO_2 + H_2O$$

nitrate nitrogen gas

It's worth noting that, bacteria would normally tend to use oxygen in oxidizing organic matter to produce more energy when nitrate and oxygen are available. Anoxic conditions are required to proceed with denitrification. The key mechanisms applied in wastewater treatment include synthesis, nitrification, ammonification, and denitrification procedures for nitrogen control and/or elimination (U.S. Environmental Protection Agency, 1993) [202].

1.2.2 Wastewater Treatment Technologies for Nelimination

There are two, most common forms of technology employed for nitrification and denitrification as following: attached growth and suspended growth, this thesis will provide a description only attached growth, particularly the BAFs.

1.2.2.1 Attached Growth

The term 'attached growth' describes a form of wastewater treatment where the wastewater influent flows past a support media upon which a biofilm develops and is often referred to as a 'biofilter'—this is shown schematically in (Figure 1.2). The support material may be stones, gravel, sand, wood, activated carbon, or plastic. Media would be arranged in an orderly, uniform fashion, or randomly packed. The wastewater usually flows by gravity as a free surface stream. As the wastewater passes over the biofilms, diffusion gradients within the film allow NH₄⁺, NO₃⁻ and dissolved organics to pass into the biofilm. Any waste products are diffused outwards and carried away by the coming stream of wastewater (Briers, 2010) [25].

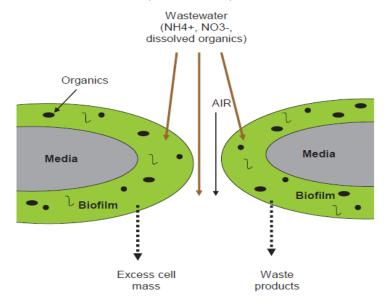


Figure 1.2. Schematic of treatment and flow in associated growth media filters (Jantrania & Gross, 2006) [91].

Within attached growth reactors, the bacteria exist in a complex aggregation called a biofilm attached to the media's surface. The oxygen gradient within the biofilm will provide different environments for bacterial growth. Dissolved oxygen will typically only penetrate 100-200 µm of a biofilm resulting in anaerobic, anoxic, and aerobic layers as illustratedin (Figure 1.3) (Biesterfeld, Farmer, Russell, & Figueroa, 2003) [20]. The layers pose some significant problems to nitrifiers that are located within the biofilm's innermost region, as it is the regular case, will have insufficient dissolved oxygen levels and denitrifiers may not have sufficient organic carbon available. Research has shown that in suspended growth and fixed film system, an oxygen gradient exists within the biofilm/bacterial floc which may consist of an aerobic outer layer and an anoxic sublayer so that aerobic processes and denitrification may occur simultaneously. Thinner biofilms are more advantageous as they ensure less diffusion with a greater surface area available with wastewater(Timberlake, Strand, & Williamson, 1988) [195].

Waste products, produced by metabolic processes occurring in the biofilm are diffused outwards and carried away by the wastewater stream. A naturally occurring replenishment system of the bacteria within the biofilm is encouraged by 'sloughing'- the increasing thickness of the biofilm whichwill be finally unable to support the outer bacterial layers which detach and are washed out with wastewater(Briers, 2010) [25].

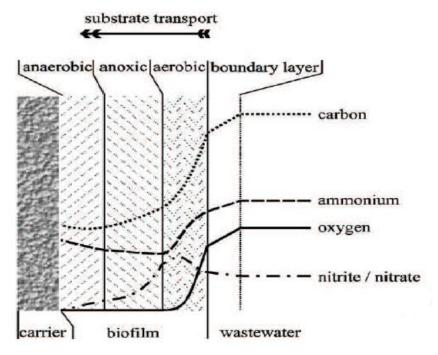


Figure 1.3. Representation of a biofilm structure under conventional aeration conditions (Rabiger:, 2005) [153].

1.2.2.2 Biological Aerated Filter Systems (BAFs)

BAFs were established in Europe then applied worldwide as a newsystem for treating wastewater forits benefitsover other treatment systems (Allan, Leopoldo, & Tom, 1998) [11]. The term BAFsformed from a mixture of air and the bacterial filtration action. The BAFsnormally (Figure 1.4) include a medium which treats nitrogenous and carbonaceous matter by medium-fixed biomass and collecting the suspended solids in the media (Ha, Ong, & Surampalli, 2010) [69].

Conventionally, the BAFs are waterlogged media wastewater treating reactors, which mixboth separating biomass by depth-filtration and aerobic biological treatment (Mendoza-Espinosa

&Stephenson, 1999) [123]. It utilises granular media as a support for microbial biofilms and also serve as a depth filter.BAFs are a low-impact alternative to the standard aerobic process, which would be functioned at higherBOD loadings compared to activated sludge and trickling filters. In one-unit process of BAFs, solids filtration, elimination of carbonaceous BOD and nitrification would be utilized(Clark, Stephenson, & Pearce, 1997) [41]. BAFs are available in two configurations: up-or down-flow. For the up-flow BAFs, the influent is launched at the bottom of BAFs and flows co-current with air while for the down-flow BAFs, the influent is fed at top of BAFs and flows against current to air (Tan, 2007) [191]. BAFs would be applied to achieve instantaneous elimination of carbonaceous BOD and solids (secondary treatment), ammonia and solids (tertiary treatment), and/or removal of solids, carbonaceous BOD, and ammonia (combined secondary and tertiary treatment) in one operating unit. Moreover, additional design alterations allow for denitrification removal. (Condren:, 1990) [42].Through adding biomass to media, BAFs may also be used to treat refractory wastewater, e.g. textile and oil field wastewater(Chang, Hong, & Park, 2002; Zhao, Wang, & Ye, 2006) [35], [226].

1.2.2.2.1 BAFs Process Description

The BAFs method is a three-phase submerged attached growth proceed: Solid phase is the media that supports biofilm formation, liquid phase is the influent settled sewage, and Gas phase is the air provided to the system (Mendoza-Espinosa & Stephenson, 1999) [123]. The sewage moves in the system up or down. Wastewater treatment occurs because of concurrent physical and biological procedures that occur as drainage comes with the biofilm-coated media(Boardman, Novak, & Zhang, 2015) [22].

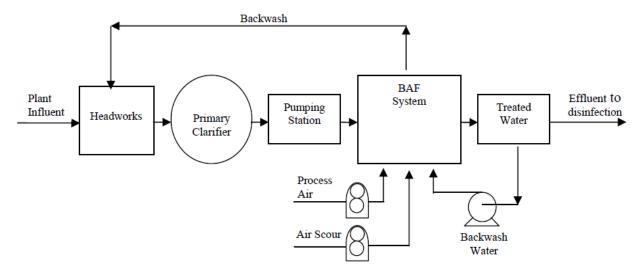


Figure 1.4. Typical of the BAFs for wastewater treatment (Rajan, 2010) [155].

The nitrogen conversion and removal in BAFs that treat municipal wastewater is under interest in the current study. According to the literature, BAFs could produce a better-quality effluent, less land, compared to other systems e.g. activated sludge and trickling filter procedures, in one- operation unit of BAFs, nitrification, carbonaceous BOD elimination, and solids filtration can be utilized.

1.3 Problem Statement of the Research

Filtration media-based bed types of choice are crucial in BAFs' procedure and design to attain effluent quality. According to the literature, a variety of filtration media types use an attached growth zone in the BAFs. Nevertheless, several filtration media are manufactured and proprietary. The usage of readily accessible media has been tested but has not been widely used. In this way, thus the performance of the 0.78±0.60 mm activated carbon bed, 0.95±0.58 mm sand bed, and 3.28±2.14 mm ceramic bed as attached growth zones for the creation of local filtration media beds, low cost, for improving the effluent quality.

1.4 Scope of the Research

The scope of the current research was to undertake investigations of the nitrogen conversion and removal by three identical pilot-scale downflow of BAFs using 0.78±0.60mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed as biomass support as attached growth zones in the treatment of municipal wastewater.

1.5 Aim and Objectives of the Study

Mainly, the present study is designed to respond to the following question of the study: What is the efficacy of 0.78±0.60 mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed used as attached growth zones in downflow of BAFs for conversion and N-elimination? this allows defining the main goal of the thesis was nitrogen conversion and removal by bio-aerated filtration (BAFs), as well as various goals consequent from this main objective as follows:

- 1. Impact of three different filtration media types on the elimination of ammonium-nitrogen in BAFs.
- 2. Nitrification & denitrification procedure by three filtration media types in BAFs.
- 3. Combined COD and ammonium-nitrogen elimination a BAFs comparison between three different filtration media types.
- 4. The ratio sCOD/TKN Influence on the nitrification process comparison between three filtration media types in BAFs.
- 5. Effect of DO, Alkalinity, and pH on nitrification by three filtration media types in BAFs.

2 Nitrogen Removal - Current Status

2.1 General

In this chapter, overview of current status-nitrogen removal and BAFs technology including, focusing on biological nitrogen removal and the recent progress and several other pathways available to eliminate nitrogen from wastewater. Also, this chapter presented the progress of BAFs and the proprietary system, description mechanism of the nitrogen conversion and removal in the BAFs including nitrification and denitrification principles are presented.

2.2 Biological Nitrogen Removal

Nitrate, Ammonia, particulate, and soluble organic N make up total effluent nitrogen. Nitrification and denitrification are the biological mechanisms that mainly eliminate nitrogen (Jeyanayagam, 2005) [92].

The nitrification: *Nitrosomonas*, a kind of autotrophic bacteria, converts ammonia to nitrite during the nitrification procedure. Another autotrophic bacteria community, *Nitrobacter* is the most frequent, then oxidizes nitrite to nitrate.

The denitrification: biological conversion of nitrate to nitric oxide, nitrous oxide, and nitrogen gas is known as denitrification. Denitrification is possible in both heterotrophic and autotrophic bacteria. *Pseudomonas* species are the widely spread denitrifying bacteria, which can denitrify hydrogen, methanol, sugars, organic acids, alcohols, benzoates, and other aromatic compounds (Metcalf & Eddy, 2003) [125].

2.2.1 Recent Developments in Biological Nitrogen Removal

Recent research into microbial nitrogen removal has revealed several other pathways available to eliminate nitrogen from wastewater. These include partial nitrification (the SHARON process), anaerobic ammonium oxidation (ANAMMOX), entirely autotrophic N elimination over nitrite (CANON), and NO_x procedure. Figure 2.1 shows, for comparative purposes (a) the classical nitrogen cycle and (b) an overall nitrogen web, integrating current findings and novel pathways in the N- cycle.

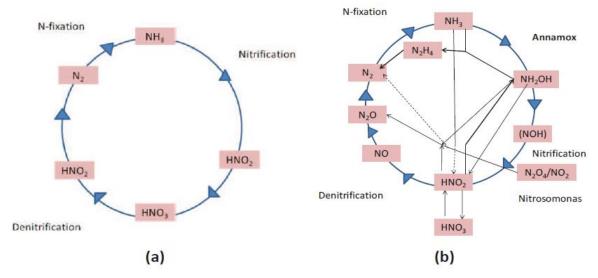


Figure 2.1.The recently re-classified nitrogen cycle. (a) shows the classical nitrogen cycle whilst (b) shows the general N web integrating potential biochemical pathways (Ahn, 2006) [5].

Partial nitrification involves inhibiting subsequent nitrite to nitrate conversion. At sufficiently high temperatures – above 26°C – the SHARON method utilises the varying growth rates oxidizers(nitrite and ammonia)(Schmidt, et al., 2003) [180]. A hydraulic retention time is then set among the specific growth-rates of the nitrite and ammonia oxidizers which results in the nitrite oxidizers being washed from the reactor before conversion to nitrate. This procedure would be integrated with annamox procedure or conventional denitrification to reduce the nitrite to dinitrogen gas.

The annamox pathway of microbial nitrogen transformation was recently discovered by (Mulder, Van der Graaf, Robertson, & Kuenen, 1995) [127]. The process is a lithoautotrophic biological conversion reaction where the *Planctomycete* group of bacteria consumes ammonia in the lack of oxygen. Ammonia is oxidised producing dinitrogen gas, using nitrite as the electron-acceptor and CO₂ as the C-source. It must beincompletely (55-60 % of ammonium) pre-oxidized to form nitrite, not nitrate, prior tonourishing to annamox process (Ahn, 2006) [5]. One concern with the anammox system is the longer start-up times due to slow-growing *planctomycetes* (Schmidt, et al., 2003) [180].

In a single aerated reactor, the CANON process incorporates partial nitrification and anammox. Nitrifiers convert ammonia to nitrite, absorb oxygen, and thus forming anoxic environments for anammox (Ahn, 2006) [5].

The NO_x procedures featured by the regulation and simulation denitrification activities of *Nitrosomonas*-like microorganisms. This is achieved by addition of trace quantity of N-oxides to promote simultaneous nitrification and denitrification and produce dinitrogen as an end product (Zart & Bock, 1998) [222]. The process can occur even under fully aerobic conditions.

The above technologies are all currently in the development stage with the first full-scale reactors only recently being tested (Abma, et al., 2007; Joss, et al., 2009; Van der Star, et al., 2007; Wett, 2006) [2], [96], [203], [215]. (Li, Sun, & Xu, 2008) [109], considering the fact that gradual establishment of annamox reactors in some countries over the last few years, the annamox process is only primarily used to extract ammonium from sludge digester effluents and landfill leachate.

Since the successful implementation of SHARON (Single reactor system for High Ammonia Removal Over Nitrite process) in practise, nitrification and denitrification via nitrite technology have garnered increasing interest. The incomplete nitrification mechanism works because nitrite serves as a linkage between denitrification and nitrification stages: incomplete nitrification is accompanied by nitrite denitrification (Figure 2.2)(Ferhan, 1996; Fdz-Polanco, Villaverde, & Garcia, 1996) [62], [58].

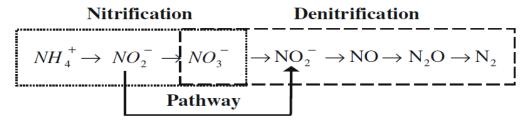


Figure 2.2. Biological nitrification-denitrification by nitrite mechanism (Peng & Zhu, 2006) [140]

In comparison to conventional nitrification and denitrification through nitrate, the following are the primary advantages of partial nitrification over total nitrification (Beccari, Marani, Ramadori, & Tandoi, 1983; Turk & Mavinic; van Kempen, Mulder, Uijterlinde, & van Loosdrecht, 2001) [16], [198], [204]:

- 25% less O₂ intake in aerobic phase equates to a 60% energy save;
- The electron donor needs is reduced by up to 40% throughout anoxic stage;
- Nitrite denitrification rates show 1.5 to 2 times greater than nitrates;
- Decrease CO₂ emission by 20%;
- 33~35% reduce sludge formation in nitrification and 55% in denitrification procedures.

While treating wastewater with elevated ammonium levels or low C/N ratios, incomplete nitrification to nitrite and nitrite denitrification was documented as theoretically feasible and economically beneficial (Surmacz-Gorska, Cichon, & Miksch, 1997; Hellinga, Schellen, Mulder, van Loosdrecht, & Heijnen) [190], [82]. The SHARON method is the earliest to successfully achieve nitrification/denitrification using nitrite as an intermediary in a commercial scale and under constant conditions (Mulder, van Loosdrecht, Hellinga, & van Kempen, 2001) [128]. Table 2.1. represent the full-scale implementation of SHARON procedure for N-elimination.

Table 2.1.SHARON process, full-scale application of for nitrogen elimination (Peng & Zhu, 2006) [140]

Full-scale plants	Capacity (pe)	Influent	N removal (%)	N load (kg N/d)	Operational year
Utrecht	400,000	Rejection water	>95	900	1997
Rotterdam	470,000	Rejection water	>95	850	1999
Zwolle	200,000	Rejection water	>95	410	2003.
Beverwijk	320,000	Rejection water and condensate	>95	1.200	2003
Garmerwolde	300,000	Filtrate and condensate	>95	2,500	2005
Hague	930,000	Centrate	>95	1,200	2005

However, SHARON's operating conditions, e.g. high temperature and ammonium, constrain its production and implementation (STOWA, 1995) [187]. Additionally, the application varies primarily from stage to phase on sludge digestion rejection water and landfill leachate water. The question of how to attain and sustain steady incomplete nitrification in different types of wastewater and under popular conditions continues to attract the attention of researchers. Until now, only a few efficient partial nitrification processes have been achieved in a sequential operation, and only a few in a continuous-flow process(Peng, Chen, & Tian, 2003; Khin & Annachhatre Ajit, 2004) [141], [100]. This study discusses and critically appraises many innovative technologies for effective partial nitrification. Additionally, the current state of the application is discussed and potential future research areas for further protocol understanding.

2.2.2 Biological Relations of Ammonia-oxidizing and Nitrite-oxidizing Bacteria

Nitrification is a sequential biological-oxidation mechanism involving two distinct bacterial group ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB). The first stage in nitrification is oxidizing ammonia to nitrite over hydroxylamine (NH₂OH), which is carried out by AOB. The NOB group oxidises nitrite to nitrate further. Currently, no single strain of bacteria capable of directly oxidising ammonia to nitrate was identified (Radajewski, et al., 1994; Regan, Harrington, & Noguera, 2002; Lipponen, Martikainen, Vasara, Servomaa, Zacheus, & Kontro, 2004) [154], [156], [111]. The oxidation of ammonia to nitrite is a speed-determinate reaction under common conditions; however, nitrite is quickly oxidised to nitrate, and therefore nitrite is accumulated in nitrifying reactors.

Nevertheless, nitrite production is necessary in the partial nitrification procedure, and the second stage to be restricted to washout NOB and accumulate AOB (Laanbrock & Gerards, 1993) [103].

AOB and NOB coexist and prosper from their physical proximity during the nitrification phase. The near physical interaction is advantageous for energetic causes: NOB can effectively intercept the nitrite (i.e., their substrate) formed by the AOB, thus compensating for the nitrite oxidation's low energy yield. Furthermore, AOB appreciates the NOB's existence because it protects them from radioactive nitrite. Therefore, it helps in the protection versus nitrite toxicity by avoiding the aggregation or development of harmful by-products e.g. NO, which may interfere with bacterial enzymes(Stein & Arp, 1998) [185]. However, relevant experiments using comparative 16S rRNA analysis of AOB and NOB have established and present in two distinct lineages within the Proteobacteria (Head, Hiorns, Embley, McCarthy, & Saunders, 1993; Teske, Alm, Regan, Toze, Rittmann, & Stahl, 1994) [81], [194].

The main bacterial essential for ammonia and nitrite oxidation are Nitrobacter and Nitrosomonas, both chemolithoautotrophic members of the Proteobacteria genus (Wheaton, Hochheimer, Kaiser, Kronos, Libey, & Easter, 1994) [216]. The characterised autotrophic AOB are all members of the Proteobacteria subdivision and are typified by *Nitrosomonas Europaea* (Figure. 2.3). The SHARON protocol is mainly performed by *Nitrosomonas Eutropha* (Logemann, Schantle, Bijvank, van Loosdrecht, Kuenen, & Jetten, 1998) [112]. Within the subdivision, these bacteria form a distinct community with an iron-oxidizing bacterium and *Rhodocycluspurpureus* (a photosynthetic bacteria), as well as methylotrophic bacteria. *Nitrosococcusoceanus* is the only exemption, since it is a marine genus in the -proteobacterial lineage. The Proteobacteria has a higher prevalence of NOB. The most extensively researched autotrophic NOB in the subclass Proteobacteria, with *Nitrobacter winogradskyi* serve as a representative group. Further chemolithoautotrophic NOB that have been identified are widespread phylogenetically in the Proteobacteria class and subdivisions (Figure 2.3).

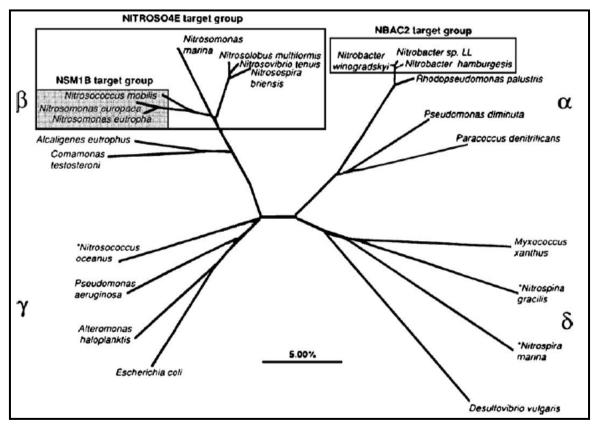


Figure 2.3. Phylogenetic map of the chemolithoautotrophicammonia- and NOB (Timothy & Edward, 1996) [196].

2.3 Biological Aerated Filters BAFs

Suspended and attached growth biological systems were applied in wastewater treatment. BAFs is an attached growth bioreactor, combining biological treatment and filtration in a single unit. BAFs would be applied as secondary or tertiary treatment units, it may also be called; Submerged Aerated Filters. It is essential to have a submerged bioreactor in which packing media possessing a higher surface area is used to maintain biomass growth. Settled wastewater flows in up-or down-flow state through the packing media. Air is supplied by means of diffusers. The high biomass concentration per unit volume makes BAFs a compact and flexible reactor as versus other wastewater treatment options (Canler & Perret, 1994; Pujol, Hamon, Kandel, & Lemmel, 1994) [29], [146]. Pollutant removal occurs due to the simultaneous filtration and biological transformation that take place as wastewater diffuse through the packing media. Thus, the selection of suitable packing media plays a major role in the design, operation, and treatment objectives achieved (Moore, Quarmby, & Stephenson, 2001) [126].

2.3.1 Flow Features in the BAFs

The BAFs' flow features are recognized by the liquid and air inputs, and the media (Le Cloirec & Martin, 1984) [104]. Simultaneously, reactor hydrodynamics has an effect on the physiological and physical composition of biofilms (Wilderer, Cunningham, & Schlindler, 1995) [218]. While, several scientists have discovered that BAFs act like an entirely mixed system (Costa Reis & Sant'Anna, 1985)[43], because of either channeling and back-mixing (Boller, Gujer, & Tsuchi, 1994) [23], generally, the flow patterns within BAFs resemble plug-flow (Mann, Fitzpatrick, & Stephenson, 1995; Fdz-Polanco, Garcia, & Villaverde, 1996) [118], [58]. BAFs generally work at a water velocity of 1 to 10 m³m⁻² h⁻¹(Stensel, Brenner, Lee, Melcer, & Rakness, 1988; Paffoni, Gousailles, Rogalla, & Gilles, 1990; Vedry, Paffoni, Gousailles, & Bernard, 1994) [186], [137], [206].

Wastewater may be introduced into BAFs from either the top or bottom. Hence, BAFs can be run in either up- or down- flow way (Figure 2.4). Down-flow systems with counter-current air/water flow could have the benefit of allowing the supplied air to remain in contact with final effluent for a time-period. This is especially critical when the elimination of carbonaceous matter and ammonia is required in a single reactor. The nitrifying microorganisms in this mechanism occupy the lower filter's region collect oxygen-rich air and hence, do not experience oxygen deficiency (Gonzalez-Martinez & Wilderer, 1991) [66]. Since the drainage arrives at the tip, it avoids clogging the influent nozzles(Desbos, Rogala, Sibony, & Bourbigot, 1990) [52].

The up-flow systems with the flow of air/wastewater co-current are capable of handling higher influent flow rates than down-flow, improve filter run time while preventing the formation of air pockets, and have more efficient oxygenation so bubbles do not combine and establish their optimum surface/volume ratio. The ambient air is only in contacted and associated with the treated-effluent in up-flow systems, the risk of odor complaints because of air depriving of volatile compounds is diminished (lida & Teranishi, 1984) [89].

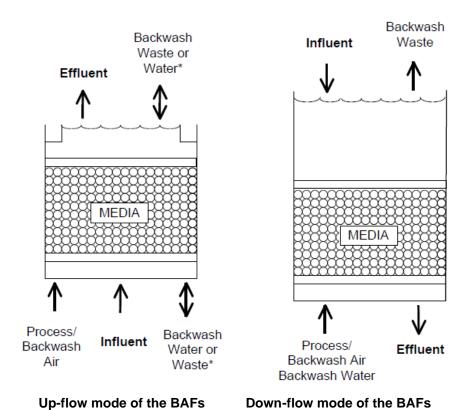


Figure 2.4.Schematic of up- and down-flow BAFs. *Up-flow BAF scan be washed by injecting backwash water either co-currently or counter currentlyto the operating direction of the flow (Mendoza-Espinosa:, 1999) [124].

2.3.2 Types of Filtration Media in the BAFs

In addition to the aeration and backwash systems, a filter material is a critical component of BAFs. The filter content has an obvious effect on the hydraulic properties and the oxygen-substrate transition rate (Mendoza-Espinosa & Stephenson, 1999) [124]. Thus, selecting an appropriate BAFs content is a vital component of the operation's and BAFs process design, since it enables the effluent output to meet the controlled norm(Tan, 2007) [191]. There are two kinds of filter media used in BAFs: floating media e.g. plastics, polyester pellets, polyurethane pellets, and sunken media e.g. zeolite, ceramics, and sand (Table 2.2). The filter media serves as a nitrifying and denitrifying biofilm accessory, providing a broad surface area per unit volume necessary for the maintenance of a high number of active biofilms and a diversity of microbial communities. Moreover, (He, Xue, & Kong, 2007) [79], stated that the media enables the reactor to function as a deep, underwater filter capable of removing suspended solids. The filter media used to achieve the necessary effluent level quality for ammonia elimination is estimated by several factors, including the media form and scale.(Kent, Fitzpatrick, & Williams, 1996) [98], noted that filter media had to be characterized in order to assess their appropriateness for the growth and development of biofilm, as well as ammonia elimination.

Table 2.2. Types of filtration media-based beds used in BAFs

Materials	Materials type	Materials size	Liquid	Reference
		mm	flow	
Sunken	expanded clay	~ 1.3	Up-flow	(Sagberg, Dauthuille, & Hamon, 1992) [175]
		~ 1.9		(Smith & Hardy, 1992) [182]
		~ 3.5		(Vedry, Paffoni, Gousailles, & Bernard, 1994) [206]
		6 - 8		(Fruhen, Bocker, Eidens, Haaf, Liebeskind, & Schmidt, 1994) [63]
	anthracite	3	Down-flow	(Terayama, Nishimura, Isono, & Hosoya, 1997) [193
	vitrified clay	3 - 4	Down-flow	(Clapp, Talarczyk, Park, & Boyle, 1994) [39]
	expanded shale	3 - 6	Down-flow	(Dillon & Thomas, 1990) [53]
		3 - 6		(Bacquet, Joret, Rogalla, & Bourbigot, 1991) [14]
		2 - 6		(Robinson, Brignal, & Smith, 1994) [160]
		2.5 - 3.5		(Tschui, Boller, Gujer, Eugster, Mader, & Stengel, 1994) [197]
	porous stone	20 - 35	Up-flow	(Costa Reis & Sant'Anna, 1985) [43]
	slag	~ 40	Down-flow	(Dee, James, Jones, Strickland, Upton, & Cooper, 1994) [48]
	granular	~ 2.8	Up-flow	(Jimenez, Capdeville, Roques, & Faup, 1987) [94]
		3 - 6	Down-flow	(Pujol, Hamon, Kandel, & Lemmel, 1994) [146]
		2.5 - 3.5	Up-flow	(Pujol, Hamon, Kandel, & Lemmel, 1994) [146]
Floating	polystyrene	3 - 3.5	Up-flow	(Tschui, Boller, Gujer, Eugster, Mader, & Stengel, 1994) [197]
		3.5		(Vedry, Paffoni, Gousailles, & Bernard, First months operation of two biofilter prototypes in the waste water plant of Acheres, 1994) [206]
		3.5		(Andersen, Bundgaard, Andersen, Hong, & Heist, 1995) [11]
		2 - 3.5		(Visvanathan & Nhien, 1995) [212]
	polypropylene	2.3 - 2.7	Up-flow	(Mann & Stephenson, 1997) [117]
	recycled plastic	5	Up-flow	(Zeghal, Nagem Nogueira, Salzer, & Rogalla, 1995) [223]
Sunken	Sand	2.00	Up-flow	(Mendoza-Espinosa:, 1999) [123]
	quartz	3.15	Down-flow	. , , , , , , , , , , , , , , , , , , ,
	gravel, lava rock, long	5 -12.5 - 15	Down-flow	(Ha J. , 2006) [70]
	plastic rings			

2.3.3 Size of the Granular Filtration Mediain the BAFs

The particle size of the treatment granular media has an impact on the effectiveness of solids elimination and usable surface-area for biofilm connection(Smith & Marsh, 1995) [183]. Application of larger media (>6 mm) results in a decreased elimination of nutrient and solids due to the occurrence of large void spaces, as well as a decline in the area available for biofilm formation(Stensel, Brenner, Lee, Melcer, & Rakness, 1988) [186]. Nonetheless, using massive media minimize backwashing needs, which lowers functioning costs (Costa Reis & Sant'Anna, 1985; Pujol, Hamon, Kandel, & Lemmel, 1994) [43], [146]. Small media (3 mm) improves filtration and provides a wide surface area for biofilm formation, but it requires more regular and intense backwashing(Robinson, Brignal, & Smith, 1994) [160]. As a result, it has been recommended that coarse media >6 mm be applied for roughing, medium media 3-6 mm for a typical application, and fine media (3 mm) for tertiary treatment and/or effluent polishing (Quickenden, Mittal, & Gros, 1992) [151].

2.3.4 Nitrogen Conversion and Removal in the BAFs

2.3.4.1 Nitrification Principles in the BAFs

Autotrophic nitrifying microorganisms evolve more slowly than heterotrophic ones and are more vulnerable to environmental factors. The volume of OM degradation affects the development of autotrophic nitrifying microorganisms. Nitrification can be expressed in the following way (Rittmann & McCarty, 2002) [158]:

Nitrosomonas reaction:

$$2NH_4^+ + 3O_2 \longrightarrow 2NO_2^- + 2H_2O + 4H^+ + New Cells$$

Nitrobacterreaction:

$$2NO_2^- + O_2 \longrightarrow 2NO_3^- + New Cells$$

Total reaction including cell synthesis and respiration of nitrifiers would be expressed as:

$$NH_4^+ + 1.83 O_2 + 1.98 HCO_3 \rightarrow 0.98 NO_3^- + 0.021C_5 H_7 O_2 N + 1.88 H_2 CO_3 + 1.04 H_2 O_3$$

As shown previously, 1g of NH₃-N needs7.14 g of alkalinity and 4.33 g O₂forming of 0.15 g-cells.

Numerous environmental conditions have an impact on how well nitrifying biofilms do in BAFs. Along with the hydraulics and nutrient/substrate transport through the biofilm, the reactor setup and airliquid mass transfer both possess an effect on nitrification. Apart from physical considerations, nitrification is influenced by dissolved nutrient, alkalinity (HCO_3^-), substrate concentrations (NO_3^- , NO_2^- , COD, NH_4^+ , and O_2), pH, poisonous material concentrations, and relative microbial diversity (heterotrophs, Nitrosomonas, Nitrobacter), biomass composition, and biofilm thickness.

2.3.4.1.1 Current Status-Nitrification in the BAFs

Nitrification is the primary method used in this procedure to strip ammonia from the water by turning it to nitrate. This involves a high aeration level to provide enough oxygen and sustain the system's energy requirements. Additionally, since nitrifying bacteria generate very low energy during nitrification, bacterial replication and growth are limited. For every kg of ammonia nitrification, only 0.06 kg of nitrifying bacteria can be formed(Gerardi, 2006). Additionally, this mechanism is limited by the fact that nitrifying bacteria are involved and replicate only in the range of 5-40°C. The maximum nitrification rate is recorded at 30°C, and it completely ceases below 5°C. A concentration of 1-5 mg d⁻¹ m⁻³free ammonia prevents oxidation, and the degree of prevention is strongly dependent on pH, ammonia concentration, temperature, DO, and the growth rate of ammonium oxidizers relative to nitrite oxidizers (Turk & Mavinic, 1989) [199].

The secondary procedure BAFs is mainly performed to eliminate carbonaceous content has been shown to utilize incomplete nitrification with ammonia of up to 1 kg NH₃-N m⁻³ d⁻¹ (Table 2.3). A minor amount of ammonia in BAFs involved in tertiary nitrification is removed through cell development, while the remainder is oxidized by autotrophic bacteria (Akunna, Bizeau, Moletta, Bernet, & Heduit, 1994; Cecen & Gonenc, 1995) [7], [32].

Table 2.3.Nitrification rates obtained in BAF used for combinedsecondary and tertiary treatment, and for tertiary treatment only.

Reactor configuration	Type of Treatment	Maximum nitrification rate (kg NH ₄ +-N m ⁻³ d ⁻¹)	Reference
Up-flow	combined	1.0	(Rogalla & Bourbigot, 1990) [161]
Up-flow	combined	0.19	(Carrand, Capon, Rasconi, & Brenner, 1990) [30]
Down-flow	tertiary	0.39-0.84 ‡ (at 22°C)	(Rogalla & Payraudeau, Tertiary nitrification with fixed biomass reactors, 1988) [162]
Down-flow	tertiary	0.6 †	(Schlegel, 1988) [179]
Down-flow	tertiary	0.9 - 1.2	(Sakuma, Tanaka, & Maki, 1993) [177]
Down-flow	tertiary	0.5 (at 13°C)	(Paffoni, Gousailles, Rogalla, & Gilles, 1990) [137]
Up-flow	tertiary	0.12-0.54	(Fruhen, Bocker, Eidens, Haaf, Liebeskind, & Schmidt, 1994) [63]
Up-flow	tertiary	0.55	(Fdz-Polanco, Villaverde, & Garcia, 1994) [60]
Down-flow	tertiary	0.22	(Terayama, Nishimura, Isono, & Hosoya, 1997) [193]

 $^{^{+}}$ kg TKN m^{-3} d^{-1}

2.3.4.1.2 Effectiveness of Hydraulic and Ammonia Loading on BAFs

BAFs have been demonstrated to perform effectively at higher rates of organic and hydraulic loading than activated sludge techniques(Belgiorno, Feo, & Napoli, 2003; Hamoda, Al-Ghusain, & Al-jasem, 2004; Mendoza-Espinosa & Stephenson, 1999; Rogalla, Lamouche, Specht, & Kleiber, 1994; Peladan, Lemmel, & Pujol, 1996) [19], [73], [123], [165], [139]. Additionally, BAF systems are good at removing N and COD while maintaining a limited footprint(Rogalla & Bourbigot, 1990; Rogers, Clifford, Mulqueen, & Ballantyne, 2004; Westerman, Bicudo, & Kantardjieff, 2000)[161], [166], [216]. Table 2.4 summarises the organic carbon and ammonia elimination levels in BAFs under varying conditions from the literature.

Table 2.4.Organic C and ammonia elimination in BAFs under nitrification condition

Configuration	Ammonia	COD	Media	Media	NH ₄ +-N	COD	Reference
	loading rate (kg/m³ day)	loading rate (kg/m³ day)	type	Size (mm)	removal (%)	removal (%)	
Down-flow	0.3 - 0.48 (TKN)	3.5 - 4 (BOD)	Clay	3.4 - 4.4	74 - 80	85	(Stensel, Brenner, Lee, Melcer, & Rakness, 1988) [186]
Down-flow	0.6	3 (BOD)	Sand	3 - 6	95	65	(Paffoni, Gousailles, Rogalla, & Gilles, 1990) [137]
Down-flow	-	3 - 5 (BOD)	Anthracite	_	_	83 (BOD)	(Adachi & Fuchu, 1991) [4]
Up-flow and Down-flow	0.36	3.5 - 11.9	Expanded clay	2.7 - 6	68 - 80 (TKN)	69.8-78.8	(Canler & Perret, 1994) [29]
Up-flow	1.5	10	Clay	3 - 6	_	_	(Pujol, Hamon, Kandel, & Lemmel, 1994) [146]
Up-flow and Down-flow	0.4 - 1.5	_	Granular Slate	2.5 - 3.5	-	-	(Boller, Gujer, & Tsuchi, Parameters affecting nitrifying biofilm reactors, 1994) [23]
Down-flow	0.4	2 (BOD)	Sand	_			(Rogalla, Lamouche, Specht, & Kleiber, 1994) [165]
Up-flow	2.7	_	Clay	_	90	_	(Peladan, Lemmel, & Pujol, 1996) [139]
Up-flow	1	2	Clay	-	51	94	(Pujol, Lemmel, & Gousailles, 1998) [147]

[‡] kg NH₄-N m⁻³ media d⁻¹

2.3.4.1.3 The Ratio Carbon/Nitrogen - Influence on the Nitrification

The ratio Carbon/Nitrogen (C/N ratio) in the wastewater, was shown to have an effect on the delivery of nitrifiers and heterotrophs by biological filters (Fdz-Polanco, Méndez, Uruena, Villaverde, & Garcia, 2000; Li, Luan, Zhu, Gong, & Cangcong, 2002) [59], [110]. It is critical to consider the impact of the C/N ratio on N elimination from wastewater while optimizing the biofilm reactor.

Furthermore, the pre-treatment of raw wastewater for SS elimination can result in loss of OM, lowering the wastewater's C/N ratio. Adequate C should be available to denitrify nitrates during the nitrogen elimination process entirely (Osorio & Hontoria, 2002) [136]. In a large-scale analysis of predenitrification (Water Pollution Control Federation, 1983) [215], partial denitrification was demonstrated in the COD/TKN ratio less than 15.

Municipal wastewater is often poor in C/N, resulting in inefficient N-recovery in municipal treatment plants (Zhu, Zhang, Quan, & Li, 2015) [228]. Traditionally, a C source (e.g., methanol, fructose, glucose, or sodium acetate) is added to supply denitrification with electron donors, which is a cost-cutting procedure. (Chu & Wang, 2011; Strong, McDonald, & Gapes, 2011) [37], [188]. SND can be an effective method of treating wastewater with a low C/N ratio by enhancing anaerobic intracellular C storage (Wang, Wang, Xue, Li, Dai, & Peng, 2015) [214]. Due to the fact that excessive polyhydroxyalkanoate intake was prevented in the aerobic system, denitrification in the subsequent anoxic phase was further effective(Zhao, et al., 2018) [225].

2.3.4.2 Denitrification principles in the BAFs

Heterotrophic denitrifying species synthesize nitrate reductase during the process of reducing nitrate to nitrogen gas, the end result of denitrification. Instantaneously, NO₃-N is formed and used in the production of new cells, with nitrate serving as an electron acceptor. Denitrification is highly depending on carbon supply. Denitrification is a process in which heterotrophic microorganisms reduce nitrate in the following sequence:

$$NO_3^- \longrightarrow NO_2^- \longrightarrow NO \longrightarrow N_2O \longrightarrow N_2$$

(Rittmann & McCarty, 2002) established the balanced denitrification reaction using methanol as a carbon source:

$$6 \text{ NO}_3^- + 5 \text{ CH}_3\text{OH} \longrightarrow 3 \text{ N}_2 + 5 \text{ CO}_2 + 7 \text{ H}_2\text{O} + 6 \text{ OH}$$

The general reaction with denitrifier formation and respiration (cell formula = $C_5H_7O_2N$) can be expressed as:

$$NO_3^- + 1.08 CH_3OH + 0.24 H_2CO_3 \longrightarrow 0.06C_5H_7O_2N + HCO_3^- + 1.68H_2O + 0.47N_2$$

One gram of NO₃⁻-N during denitrification and denitrifieruses 2.47 g of methanol, producing about 3.57 g of alkalinity, and formation of 0.45 g-cell.

2.3.4.2.1 Current Status-Denitrification in the BAFs

It has been shown that biological filter systems can work effectively at higher nitrate and organic loading rates over suspended growth schemes (Abu-Ghararah, 1996; Nogueira, Zeghal, & Molleta, 1998; Rabah & Dahab, 2004) [3], [131], [152]. The effects of the packed bed immersed filters, and fluidized beds on denitrification are discussed and summarized in Table 2.5 for a variety of operating and construction conditions.

Table 2.5.Organic C and nitrate elimination in biological filters under denitrification condition

Configuration		COD	Media	Media	NO ₃ -N	COD	Reference
J	loading rate	loading rate	type	Size	removal	removal	
	(kg/m³/day)	(kg/m³ day)		(mm)	(%)	(%)	
Up-flow	2.5	_	Bead	0.42- 0.55	-	-	(Hermanowicz & Cheng, 1990) [84]
Up-flow	1.5	10	Light weight bead	-	75	89	(Rogalla & Bourbigot, 1990) [161]
Up-flow and Down-flow	0.36	3.5 -11.9	Expanded clay	2.7-6	68-80 (TKN)	69.8-78.8	(Canler & Perret, 1994) [29]
Up-flow	>4	10	Clay	3-6	` - ′	_	(Pujol, Hamon, Kandel, & Lemmel, 1994) [146]
Down-flow	0.7 - 1.2	_	Anthracite	3	95	_	(Hwang, Sakuma, & Tanaka, 1994) [88]
Down-flow	1	3	Plastic polymer packing	_	90	-	(Ryhiner, S0rensen, Birou, & Gros) [172]
Up-flow	_	5	Glass ring	15-25	86	95	(Chui, Terashima, Tay, & Ozaki, 1996) [38]
Down-flow	1	4.5	Clay and sand	1 - 2	-	-	(Koch & Siegrist, 1997) [101]
Up-flow	6.4	3 - 9.1	Sand	0.46	95	_	(Semon, Sadick, Palumbo, Santoro, & Keenan, 1997) [181]
Up-flow	0.6 – 1.2	20	Expanded clay	3-6	68	93	(Pujol & Tarallo, 2000)[145]
Up-flow	5.68 - 6.69	18.59 - 26.71	Glass Raschig ring	26 x 25	97.5 - 100	99.4	(Ong, Ng, Lee, & Hu, 2002) [134]

Denitrification may be greatly harmed in the BAF if the effluent has a high level of suspended solids (SS). That is why, in urban wastewater systems, BAFs are often used after primary care. (Gilmore, Husovitz, Holst, & Love, 1999) [65]. Inadequate SS treatment will reduce the efficiency of BAFs by interacting with mass O₂ and substrate transport into the biofilm (Westerman, Bicudo, & Kantardjieff, 2000) [216]. Denitrification is accelerated during the nitrogen reduction process when there is enough carbon present (Henze, 1991) [83].

(Han, Yun, & Kim, 2001) [74], The features of an up-flow biological aerated filter's autotrophic nitrification and denitrification were investigated. Their goal was to determine the nitrification efficacy of a BAF with porous media and the probability of concurrent nitrification and denitrification without including organic-C when O₂ was scarce. The nitrification of wastewater was carried out using laboratory-scale up-flow BAFs with porous polyurethane-based media. Aerobic and anaerobic areas were present in the macropores. 1.8 kg NH₄+/m³/d was added to the wastewater. They discovered that when ammonia loading and nitrification rate were increased, DO level improved with height (from the base). This was due to the fact that air and wastewater were added simultaneously at the bottom. At the bottom, the average loading values were 0.7-1.0, 1.4-1.8, and 1.8-2.5 mg/d/m³,

respectively, at superficial air speed of 0.1, 0.2, and 0.3 cm s⁻¹. They attained a nitrification efficiency of 90%, relative to the 80–90% attained by (Pujol, Hamon, Kandel, & Lemmel, 1994) [146]. They discovered that N was out of equilibrium in the method and attempted to quantify it by measuring the overall mass balance of NH₄⁺-N, NO₂ -N, NO₃ -N, and N transferred to biomass. At low loadings, the expected loss was low, but about 40% of NH₄⁺-N was lost at high loadings. They speculated that the system was undergoing simultaneous nitrification and denitrification in anoxic microzones in the middle of the sludge floc or in the internal part of the biofilm adjacent to the media. Absolute ammonia is measured twice a week at the LRWRP, providing about 120 data points for 2009.A total of 76% ammonia elimination was achieved, at air flow rate of 1300 m³ h⁻¹ to 1700 m³ h⁻¹ per cell (Rajan, 2010) [155].

2.3.4.2.2 Factors Affecting Denitrification in the BAFs

Denitrifying biofilms' efficiency is influenced by the temperature of the wastewater. Denitrification can occur between 35 and 55 °C, but the reaction rate slows significantly at lower temperatures. According to (Lee, Ong, & Ng, 2004) [106], related growth is less temperature sensitive than suspended growth ones. DO levels of 1-2 mg- O_2 L⁻¹ have a negligible effect on attached growth processes, while 0.5 mg- O_2 L⁻¹ will inhibit suspended growth. According to (Hailing & Jorgensen, 1993) the optimal pH range for denitrification is between 7.0-7.5. For denitrifying reactors, the hydraulic loadings and nutrient transportation circumstances to the biofilm surface, the reactor structure, and the basic reactor state are also critical. As previously mentioned, denitrification efficiency is influenced by the concentration of electron donor NO_3^- and the C/N ratio. They can be run at a lower C/N ratio than suspended growth because the longer SRTs result in less carbon needed for cell synthesis and a lower C/N ratio. (Hailing & Jorgensen, 1993) [71].

3 Materials and Methods

3.1 General

This chapter provides an explanation of the experimental study in nitrogen conversion and removal by biological aerated filtration. Nevertheless, the exact materials and methods applied, for instance, developed three identical pilot-scale downflow of BAFs and start-up for simulation the nitrogen conversion and removal. The effluent samples were collected at the end of the pilot-scale downflow of BAFs run for laboratory analysis.

3.2 Research Framework

The aim of this research was to undertake investigations of the nitrogen conversion and elimination by biological aerated filtration, general description of the research framework applied to achieve the objectives (Figure 3.1).

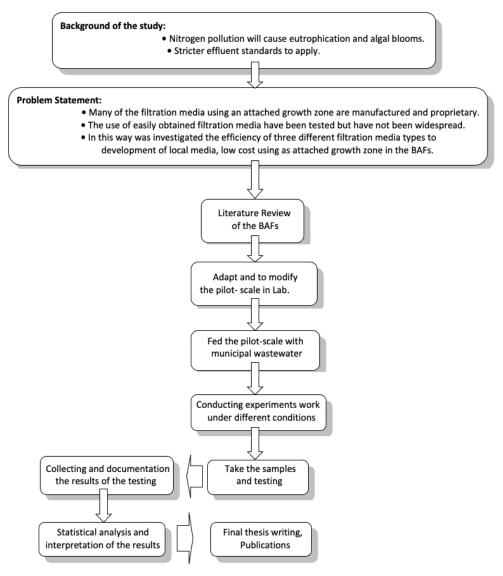


Figure 3.1. The research framework.

3.3 Experimental Set-up

Three identical pilot-scale downflow of BAFs were operated in the laboratory of the Technical University of Civil Engineering of Bucharest at Colentina. The whole experimental period 33 days including fed of the pilot-scale downflow of BAFs with raw municipal wastewater and resolved the operating problems. Was the experimental period from 2nd September 2019 to 4th October 2019.

3.4 Description of the Pilot-Scale Reactors of BAFs

Three identical pilot-scale downflow of BAFs (Figure 3.2), were constructed using PVC pipes. Each pilot-scale reactor was 0.10 m internal diameter, 2.76 m height, and 0.20 m clearance at the head of the reactors to allow the influent recirculation. The total height of each pilot-scale reactor 2.96 m. The height of the filtration media bed was 1.00 m.

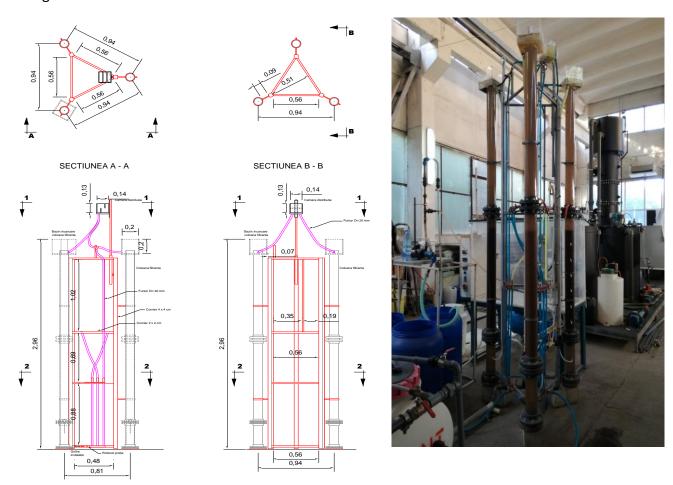


Figure 3.2. Schematic diagram and photograph of the pilot-scale downflow of BAFs.

The first pilot-scale downflow of BAFs contained 7.855 L of activated carbon-based filtration media bed, the second pilot-scale downflow of BAFs contained 7.855 L of sand-based filtration media bed, and the third pilot-scale downflow of BAFs contained 7.855 L ceramic particle-based filtration media bed, (Table 3.1) based on a working volume 65.037 L as 21.679 L in each pilot-scale of BAFs. The mean influent flowrates were 0.18 L/min, and the hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100%. While airflow was controlled by a glass VA flow-meter at the

air:liquid ratio 10:1 capacity during normal operation, the pilot-scale downflow of BAFs were backwashed every day for the whole experimental period. The pilot-scale downflow of BAFs operated at ambient air temperature ranged from 8 to 29°C with a mean of 17.93±7.27 °C.

Table 3.1. Summary description of the pilot-scale downflow of BAFs.

Characteristic	Value
constructed	using PVC pipe
number of reactors	3
internal diameter of each reactor	0.10 m
height of each reactor	2.76 m
clearance at the head	0.20 m
total height of each reactor	2.96 m
column area of the pilot-scale	2.648 m ²
filtration media-based bed	activated carbon, sand, ceramic particle
particle size	0.78±0.60 mm, 0.95±0.58 mm, 3.28±2.14 mm
total bed height of each reactor	1.00 m
volume bed of each reactor	7.855 L
mean influent flowrates	0.18 L/min
hydraulic retention time	12-hours
air : liquid ratio	10 :1
recirculation	100%
backwash	once/day

Note: The particle size shown is the mean ± standard deviation.

3.4.1 Characteristics of the Filtration Media

The size of a BAFs filtration media-based bed has a strong impact on the performance. The media-based bed significantly influences the hydraulic features and the O₂-substrate mass transfer. Consequently, different sized materials have been suggested (Mendoza-Espinosa & Stephenson, 1999) [123], where that large filtration media (>6 mm)recommended to be applied for roughing, intermediary size (3-6 mm) for overall treatment, and fine size (<3 mm) for effluent polishing the nitrogen and/or for tertiary processing.

From the (Table 3.2) it can be seen that different filtration media-based beds, used as attached growth zone in the pilot-scale downflow of BAFs, has a fine particle size of 0.78±0.60 mm, 0.95±0.58 mm, and an intermediate particle size of 3.28±2.14 mm, respectively for activated carbon-based media bed, sand-based media bed, and ceramic particle-based media bed. Generally, all three media-based beds used in the pilot-scale downflow of BAFs, are resistant to attrition, chemically inert, and have different particle sizes (Figure 3.3), it can be considered acceptable for use as biomass support as attached growth zone.

Table 3.2. Properties of media-based beds used in the pilot-scale downflow of BAFs.

Parameter	activated carbon-based media bed	sand-based media bed	ceramic particle-based media bed
Particle size	0.78±0.60 mm	0.95±0.58 mm	3.28±2.14 mm
Volume bed	7.855 L	7.855 L	7.855 L
Materials bed height	1.00 m	1.00 m	1.00 m

Note: The particle size shown is the mean \pm standard deviation.

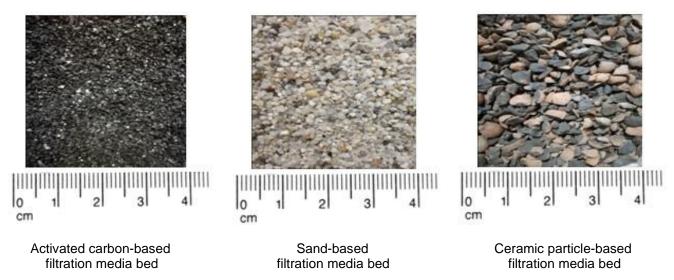


Figure 3.3. View of filtration media-based beds used in the pilot-scale downflow of BAFs.

3.5 Sampling and Laboratory Analyses

During the experimental period, composite samples were picked up of influent and effluent from the pilot-scale downflow of BAFs for laboratory analysis that took place on a daily basis five times weekly. Throughout this period the effluent samples were collected at the end of the pilot-scale downflow of BAFs run (Figure 3.4). The samples from influent and effluent were evaluated for the following quality indicators: soluble chemical oxygen demand sCOD, total chemical oxygen demand tCOD, suspended solids SS, nitrate-nitrogen NO₃-N, nitrite-nitrogen NO₂-N, ammonium-nitrogen NH₄+-N, total Kjeldahl Nitrogen TKN, total Nitrogen TN, dissolved oxygen (DO), alkalinity as CaCO₃, pH, and temperature °C. Analyses of the samples were conducted based on ISO standard and methods of (American Public Health Association, 2017). The influent municipal wastewater characteristics are shown in the (Table 3.3) and the mean daily volumetric loading applied characteristics (Table 3.4).

The pilot-scale downflow of BAFs was fed with raw municipal wastewater. Therefore, the influent concentrations and volumetric loading applied during the experimental period under different operating conditions, and the data provided in Tables 3.3, and 3.4 are the mean run of 33-day.

Table 3.3.Influent municipal wastewater characteristics during a period of testing conditions.

Parameter	Concentration		
tCOD	63.25±28.62 mg/L		
sCOD	41.50±12.86 mg/L		
suspended solids SS	59.50±48.59 mg/L		
ammonium-nitrogen NH ₄ +-N	3.16±2.46 mg/L		
nitrite-nitrogen NO ₂ N	2.46±2.07 mg/L		
nitrate-nitrogen NO₃N	1.99±1.76 mg/L		
total Kjeldahl Nitrogen TKN	10.46±6.74 mg/L		
potential hydrogen pH	7.22±0.20		
dissolved oxygen DO	3.45±1.03 mg/L		
alkalinity as CaCO₃	207.25±8.97 mg/L		
sample temperature	17.65 ±1.02 °C		

Note: *The concentrations shown are the mean ± standard deviation.

^{*}data shown is the mean running of 33-day.









Figure 3.4. View of the experimental work and analysis of the samples.

Table 3.4. Characteristics of the mean daily volumetric loading appliedduring the experimental period.

Parameter	volumetric loading applied		
total chemical oxygen demand tCOD	0.043404±0.01 kg/m ³ .d		
soluble chemical oxygen demands COD	0.028479±0.00 kg/m ³ .d		
suspended solids SS	0.093154±0.17 kg/m ³ .d		
ammonium-nitrogen NH ₄ +-N	0.002169±0.00 kg/m ³ .d		
total Kjeldahl Nitrogen TKN	0.007178±0.00 kg/m ³ .d		

Note: *The value of volumetric loading applied shown are the mean ± standard deviation.

The stoichiometry sCOD/TKN ratio describes the amount of organic carbon required to remove nitrate from the wastewater. Recognizing how the sCOD/TKN ratio affects nitrogen elimination from

^{*}data shown is the mean running of 33-day.

wastewater is critical for maximizing BAFs. Table 3.5 showing three different sCOD/TKN ratios (2.8, 4.0, 4.5), during three different runs.

Table 3.5. Influent sCOD and TKN versus sCOD/TKN ratio.

Influent parameter	Run 25 sCOD/TKN = 4.0	Run 29 sCOD/TKN = 4.5	Run 30 sCOD/TKN = 2.8
sCOD mg/L	38.30	29.50	70.50
TKN mg/L	9.50	6.50	25.00

3.6 Calculation of the Parameters

3.6.1 Hydraulic Load

$$Hydraulic\ Load = \frac{Flow\ rate}{Area} \tag{1}$$

Where:

Hydraulic Load = Hydraulic load $(m^3/m^2/h)$

Flow rate = Influent flow rate m³/h;

Area = Column's cross section area of the pilot-scale reactors of BAFs (m²).

3.6.2 Nitrification Performance

The nitrification performance was calculated according to the following equation:

$$\gamma_v = \frac{Q_{in}(S_{in} - S_{out})}{V_b} \tag{2}$$

Where:

 $\gamma v =$ ammonium-nitrogen removal rate, g NH₄+-N/m³ filter media/day;

 Q_{in} = influent flowrate m³/day;

 S_{in} = influent NH₄⁺-N level (mg/L);

 S_{out} = steady-state effluent NH₄⁺-N level (mg/L);

 V_b = volume of filter media (m³).

3.6.3 Efficiencies of the Various Parameters

The efficiencies of the various parameters were calculated as presented as follows the equation:

$$\eta = \frac{c_r - c_f}{c_r} \times 100 \tag{3}$$

Where:

 η = removal or reduction efficiency in %;

 C_r = the level of the influent municipal wastewater.

 C_f = the concentration of the effluent municipal wastewater.

4 Results and Discussions

4.1 General

This chapter presents the "results and discussion" of the data of experiments during the monitoring period provided a comparison between three different filtration media types used as attached growth zones in the treatment of municipal wastewater, for N conversion and removal by biological aerated filtration BAFs.

The general aim was to investigate the performance of nitrogen conversion and removal by biological aerated filtration BAFs. The laboratory experiments were on three identical pilot-scale downflow of BAFs (See chapter, 3 Table 3.1), using three different filtration media types, 0.78±0.60 mm activated carbon-based media bed, 0.95±0.58 mm sand-based media bed, and 3.28±2.14 mm ceramic particle-based media bed (see chapter 3 Table 3.2), to develop of local filtration media bed as biomass support as attached growth zone, low cost, to improving the effluent quality of the municipal wastewater before discharge to the aquatic environment.

The nitrification and denitrification were first observed on day 24 where all three pilot-scale downflow BAFs have achieved steady-state conditions, where the number of microorganisms increased, nitrification dominated. All three filtration media-based bed performed extremely well in the nitrogen conversion and removal, as showing in figure 4.1.

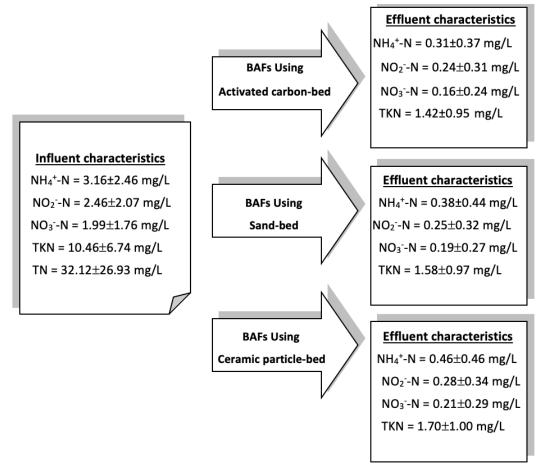


Figure 4.1.Characteristics of the nitrification and denitrification process from the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed compared with the influent municipal wastewater

Meanwhile, the results of this research showed that all the three-filtration media-based beds used in the pilot-scale downflow of BAFs were able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS, as showing in figure 4.2. Down-flow systems have the advantage of keeping the air supplied in contact with the final effluent for a longer period of time, which is critical when carbonaceous matter and ammonia elimination are needed in the same reactor. In this mode of action, the nitrifying microorganisms that occupy the lower portion of the filter collect oxygen-rich air and therefore do not experience oxygen deficiency.

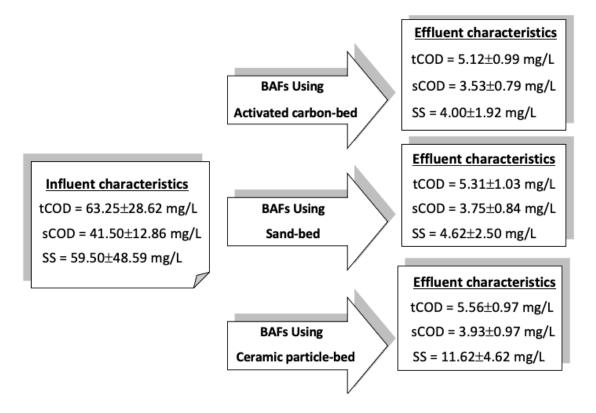


Figure 4.2.Characteristics reduction/removal of the carbonaceous from the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed compared with the influent municipal wastewater.

It can be observed the effluent from the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed compared with the influent municipal wastewater as showing in figure 4.3.



Figure 4.3.View of the effluent from the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media compared with the influent municipal wastewater.

To answer the research question this research: What is the efficiency of the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed using as an attached growth zone in the BAFs, for conversion and removal of the nitrogen. following sub-goals can be extracted from this main objective to answer the research question.

4.2 Impact of Different Filtration Media on the of Ammonium-Nitrogen Elimination in BAFs

The size of a BAFs material-based bed has a strong impact on performance. According to the experimental trials it has been investigated the efficiency of the 0.78±0.60 mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed as attached growth zones in the BAFs for ammonium-N elimination in the municipal wastewater treatment.

4.2.1 Start-up of the Experimental Trials

Throughout the duration of the experiments, all the three pilot-scale downflow BAFs were continuously fed with raw municipal wastewater containing various pollutants, e.g. ammonium-nitrogen. All the three pilot-scale downflow BAFs were operated at ambient air temperature ranged from 8 °C to 29 °C with a mean of 17.93 \pm 7.27 °C. The mean influent flowrates were 0.18 L/min, and air:liquid ratio of 10:1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and nitrogen loading rates of the three pilot-scale downflow BAFs was 0.0022 \pm 0.00 kg NH₄+-N/m³.d, 0.0072 \pm 0.00 kg TKN/m³.d, 0.0434 \pm 0.01kg tCOD/m³.d, 0.0285 \pm 0.00 kg sCOD/m³.d, and 0.0931 \pm 0.17 kg SS/m³.d.

4.2.2 Characteristics of NH₄+-N Removal forDifferent Filtration Media

During the experimental period, all the three-filtration media-based beds using in the three pilot-scale downflow BAFs showed extremely well to eliminate for NH_4^+ -N. It was investigated that the activated carbon-based filtration media bed has a slightly greater elimination efficacy compared to the sand-based filtration media bed, and ceramic particle-based filtration media bed when the mean influent of ammonium-nitrogen NH_4^+ -N load had been the same. Figure 4.4 shows the mean influent and effluents concentrations of NH_4^+ -N.

Performance of pilot-scale downflow BAFs which used a 0.78 ± 0.60 mm activated carbon-based filtration media bed. When feed with municipal wastewater contains 3.16 ± 2.46 mg NH₄⁺-N/L, with recirculation of the influent 100%, at HRT 12-hours, where the mean final effluent concentration was 0.31 ± 0.37 mg NH₄⁺-N/L, the activated carbon-based filtration media bed, was able to nitrify 0.24 ± 0.21 Kg NH₄⁺-N/m³.d, (Figure 4.5). Consistent with a mean removal efficiency of 90.11% as shown in figure 4.6.

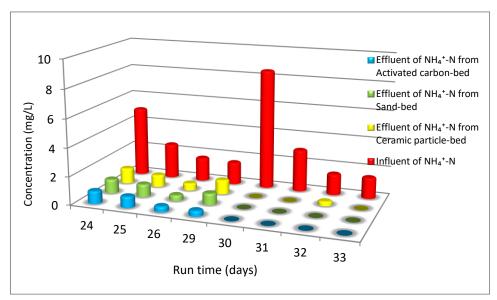


Figure 4.4. The influent and effluent concentration of NH₄+-N comparison.

Performance of the pilot-scale downflow BAFs which used a 0.95 ± 0.58 mm sand-based filtration media bed. Where feed with ammonium-nitrogen NH₄⁺-N of 3.16 ± 2.46 mg/L, with recirculation of the influent 100%, at HRT 12-hours. Where the mean final effluent level of NH₄⁺-N was 0.38 ± 0.44 mg/L, the sand-based filtration media bed, was able to nitrify 0.24 ± 0.22 Kg NH₄⁺-N/m³.d, (Figure 4.5). Corresponding to a mean removal efficiency of 87.74% as shown in figure 4.6.

Performance of the pilot-scale downflow BAFs which used a 3.28 ± 2.14 mm ceramic particle-based filtration media bed. Where feed with ammonium-nitrogen NH₄⁺-N of 3.16 ± 2.46 mg/L as well as with recirculation of the influent 100%, at HRT 12-hours, the mean final effluent level of NH₄⁺-N was 0.46 ± 0.46 mg/L, the ceramic particle-based filtration media bed was able to nitrify 0.23 ± 0.22 Kg NH₄⁺-N/m³.d, (Figure 4.5). Corresponding to a mean removal efficiency of 85.17% as shown in figure 4.6.

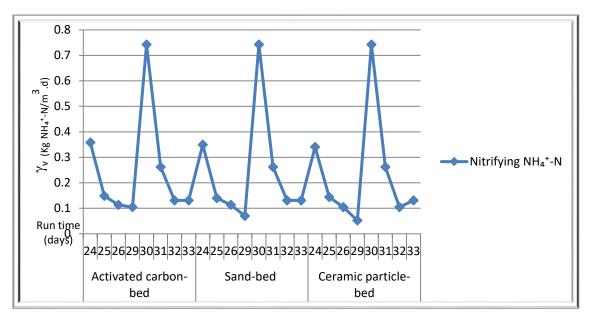


Figure 4.5. Mass removal of NH₄+-N comparison.

In figure 4.5 shows the mean mass of NH₄⁺-N ammonium-nitrogen removal in three pilot-scale downflow BAFs using three different filtration media types, 0.78±0.60 mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed with recirculation of the influent 100%, at HRT 12-hours. The experimental results indicate that the activated carbon-based filtration media bed has better capability than the sand-based filtration media bed and ceramic particle-based filtration media bed in the nitrification process. However, all three filtration media used in the pilot-scale downflow BAFs were able to reach the mean final effluent level of ammonium-N <0.50 mg L⁻¹. (Delatolla, Berk, & Tufenkji, 2008; Hussain, Aziz, & Isa, 2007), where it was discovered that the ammonia-N elimination efficacy of zeolite and carbonate BAFs was not only greater but also more consistent, than that of ceramic particle BAFs. The findings show that zeolite and carbonate media are more capable of withstanding shock loads than ceramic particles, owing to the adsorption and ion exchange properties of zeolite. (Delatolla, Berk, & Tufenkji, 2008), and the higher-porosity (Hussain, Aziz, & Isa, 2007) and alkalinity supply of carbonate (Qiu, Zhang, Wang, & Du, 2010).

4.2.3 Consumption Characteristics Related to TKN, sCOD, pH and Alkalinity

During the experiments it was recirculated the influent 100%, at HRT 12-hours. For the pilot-scale downflow BAFs used a 0.78 ± 0.60 mm activated carbon-based filtration media bed, the TKN, and sCOD concentrations decreased from 10.46 ± 6.74 mg TKN/L to 1.42 ± 0.95 mg TKN/L, and from 41.50 ± 12.86 mg sCOD/L to 3.53 ± 0.79 mg sCOD/L, with removal efficiency has reached 86.37%, and 91.47%, respectively for TKN and sCOD, as shown in figure 4.6.

For the pilot-scale downflow, BAFs used a 0.95 ± 0.58 mm sand-based filtration media bed, the TKN and sCOD concentrations decreased from 10.46 ± 6.74 mg TKN/L to 1.58 ± 0.9 mg TKN/L, and from 41.50 ± 12.86 mg sCOD/L to 3.75 ± 0.8 mg sCOD/L, with removal efficiency has reached 84.82%, and 90.96%, respectively for TKN and sCOD, as showed in figure 4.6.

For the pilot-scale downflow BAFs which used a 3.28 ± 2.14 mm ceramic particle-based filtration media bed, TKN and sCOD concentrations decreased from 10.46 ± 6.74 mg TKN/L to 1.70 ± 1.00 mg TKN/L, and from 41.50 ± 12.86 mg sCOD/L to 3.93 ± 0.97 mg sCOD/L, with removal efficiency has reached 83.69%, and 90.51%, respectively for TKN, and sCOD, as showed in figure 4.6.

The removal efficacy was largely determined by the deposition of activated biological film in the filter media bed and the efficiency of mass transfer. The primary factor that could affect COD elimination was the variation in the surface features, unique surface area, and media morphological features (Kent, Fitzpatrick, & Williams, 1996) [98],(Tay, Chui, & Li, 2003; Lee, Kim, Lee, Yun, & Choi, 2005) [192], [105]. For example, in a pilot-scale fixed-film bioreactor device, the COD elimination rate was consistently between 80-90% at an empty bed HRT of 8 h. (Jou & Huang, 2003) [97].

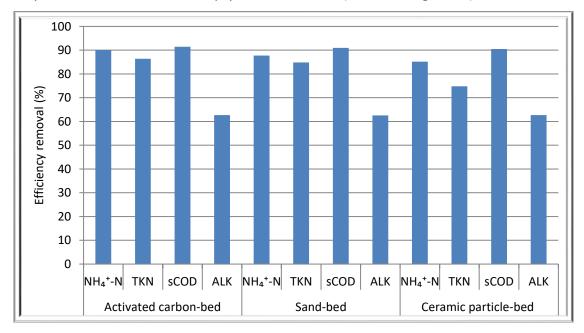


Figure 4.6. The mean removal efficiency of NH₄+-N, TKN, sCOD, and alkalinity comparison.

The performance of the pH and alkalinity according to the experimental results the mean influent pH value was 7.22±0.20 and the mean influent alkalinity was 207.25±8.97 mg CaCO₃/L, as direct evidence of nitrification, the experiments showed that more than 60 % of the influent alkalinity as calcium carbonate CaCO₃ was consumed (Figure 4.6), where the mean effluent concentration was 77.25±2.60 mg CaCO₃/L, 77.56±2.77 mg CaCO₃/L, and 77.31±2.71 mg CaCO₃/L, with pH value, was 7.21±0.20, 7.22±0. 20, and 7.22±0. 20, respectively for the pilot-scale downflow BAFs using activated carbon-based filtration media bed, the pilot-scale downflow BAFs using the sand-based filtration media bed, and the pilot-scale downflow BAFs using the ceramic particle-based filtration media bed. (Gujer & Boller, 1986) [68] indicated that an alkalinity level of at least 75 mg/L was needed to sustain the optimal nitrification rate in nitrifying biofilters used in urban wastewater treatment. The effect of alkalinity on the rate of nitrification is proportional to the pH value. According to certain sources, the pH value of wastewater should be held between 6-9 to protect organisms. (Akpor, Momba, & Okonkwo, 2008) [6]. According to an analysis on the impact of pH on nitrification efficiency in an upflow biofilter, nitrification efficiency increased linearly by 13% per unit pH rise between pH 5.0 and 8.5 (Biplob, Fatihah, Shahrom, &Ahmed, 2011) [21].

4.3 Nitrification & Denitrification Procedure by Filtration Media in BAFs

Nitrosomonas, a kind of autotrophic bacteria, oxidizes ammonia to nitrite during nitrification. Nitrite is then oxidized to nitrate by another type of autotrophic bacteria, the most prevalent of which is Nitrobacter. Denitrification is the process by which nitrate is biologically reduced to nitric oxide, nitrous oxide, and nitrogen dioxide. Denitrification is a process that can occur in both heterotrophic and autotrophic bacteria. Pseudomonas species are the most prevalent and commonly spread denitrifying bacteria, which can denitrify hydrogen, alcohols, methanol, sugars, amino acids, benzoates, and other aromatic substances.

4.3.1 Start-up of the Experimental Trials

Throughout the duration of the experiments, all the three pilot-scale downflow BAFs were continuously operated fed with raw municipal wastewater containing various pollutants, e.g. ammonium-N, nitrite-N, and nitrate-N. All the three pilot-scale downflow BAFs were operated at ambient air temperature ranged from 8 to 29° C with a mean of 17.93 ± 7.27 °C. The mean influent flowrates were 0.18 L/min, and air: liquid ratio of 10:1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and N loading rates of the three pilot-scales, downflow BAFs was $0.0022\pm0.00 \text{ kg NH}_4$ ⁺-N/m³.d, $0.0072\pm0.00 \text{ kg TKN/m}^3$.d, $0.0434\pm0.01 \text{kg tCOD/m}^3$.d, $0.0285\pm0.00 \text{ kg sCOD/m}^3$.d, and $0.0931\pm0.17 \text{ kg SS/m}^3$.d.

4.3.2 Nitrification Properties of Different Filtration Media in BAFs

To evaluate the influence of 0.78±0.60 mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed on nitrification properties of biofilter, the influent and effluent concentration of ammonium-N NH₄⁺-N at the three pilot-scale downflow BAFs shows in figure 4.4. At the aeration rate applied in the current research, DO level in the mean effluent stayed high at 6.76 ± 2.25 mg/L, 7.33 ± 1.82 mg/L, and 6.37±1.84 mg/L, respectively for the pilot-scale downflow BAFs using activated carbon-based filtration media bed, the pilot-scale downflow BAFs using the sand-based filtration media bed, and the pilot-scale downflow BAFs using the ceramic particle-based filtration media bed. Subsequently, theO₂ limitation did not prevent the functioning of all three pilot-scale downflow BAFs. (Rother, Cornel, Ante, Kleinert, & Brambach, 2002) [167], reported that in a pre-denitrification system, nitrate removal rates depend on the recycle ratio and the feed water's easily degradable carbon content that is available for denitrification. Due to the high oxygen concentrations in the nitrification zones' effluents (5 to 8 mg DO/L), a high oxygen load is recycled to the denitrification zone and easily degradable carbon is oxidized through reduction of oxygen, not nitrate. During the experiment, the pH remained between 7.21±0.20 to 7.22±0.20 which was in the optimum range for nitrification. Figure 4.1 illustrates the start-up and steady-state efficiency of three distinct filtration media-based beds. Nitrogen elimination is often cost effective in biological aerated filter systems BAFs because ammonia can be nitrified to nitrite and then denitrified in a single step. Oxygenation can be reduced by 25%, and the demand for electron donors can be reduced by 40%. Additionally, denitrification concentrations are usually 1.5-2.0 times faster when nitrite ions are used than when nitrate ions are used (Abeling & Seyfried, 1992) [1].

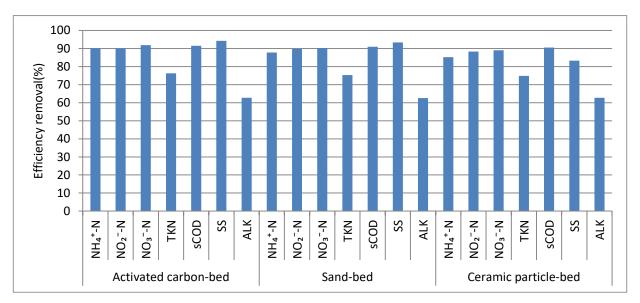


Figure 4.7. The mean removal efficiency of NH₄+-N,NO₂--N, NO₃--N, TKN, sCOD, and alkalinity comparison.

The results showed that the nitrification and denitrification were first observed on day 24 where all the three pilot-scale downflow BAFs have achieved steady-state conditions, where the number of microorganisms increased, nitrification dominated, all the three-filtration media-based beds were extremely well in the nitrogen conversion and removal. Where activated carbon-based filtration media bed has better capability than the sand-based filtration media bed, and ceramic particle-based filtration media bed in the nitrification process when the influent of ammonium-nitrogen NH₄⁺-N load had been the same. (Moore, Quarmby, & Stephenson, 2001) [126], indicated that the reactor composed of the smaller material (StarLight) exhibited improved plug flow conditions, resulting in increased nitrification and solids elimination. (Paffoni, Gousailles, Rogalla, & Gilles, 1990) [137] indicated that any more reductions in material size (StarLight) would almost certainly improve mixing to the point that the flow through the reactor no longer approximated plug flow. As a result, nitrification would degrade, as found when 3-6 mm expanded schist was used rather than 2-4 mm expanded schist. However, the experiments showed that all three-filtration media used in the pilotscale downflow BAFs were able to reach the mean final effluent level of ammonium-nitrogen less than 0.47 mg/L. Where the mean influent of the ammonium-nitrogen was 3.16±2.46 mg NH₄+-N/L, and the mean final effluent level was 0.31 ± 0.37 mg NH₄⁺-N/L, 0.38 ± 0.44 mg NH₄⁺-N/L, and 0.46 ± 0.46 mg NH₄+-N/L, respectively for the pilot-scale downflow BAFs using activated carbon-based filtration media bed, the pilot-scale downflow BAFs using the sand-based filtration media bed, and the pilotscale downflow BAFs using the ceramic particle-based filtration media bed. Where the greatest elimination efficacy of ammonium-nitrogen NH₄⁺-N was in the activated carbon-based filtration media bed was achieved 90.11%, and the mean removal efficiency slightly percentage decreased was 87.74%, 85.17% (Figure 4.7), respectively for the sand-based filtration media bed and ceramic particle-based filtration media bed. (Rogalla, Lamouche, Specht, & Kleiber, 1994) [165], reported that the elimination efficacy of NH₄⁺-N was 90%. (Paffoni, Gousailles, Rogalla, & Gilles, 1990) [137], reported that the removal efficiency of NH₄⁺-N was 95%. (Stensel, Brenner, Lee, Melcer, & Rakness, 1988) [186], reported that the elimination efficacy of NH₄⁺-N was between 74-80%. The experiments showed that the mean mass of ammonium-nitrogen removed via nitrification approximately $0.24\pm0.21 \text{ Kg NH}_4^+-\text{N/m}^3.\text{d}$, $0.24\pm0.22 \text{ Kg NH}_4^+-\text{N/m}^3.\text{d}$, and $0.23\pm0.22 \text{ Kg NH}_4^+-\text{N/m}^3.\text{d}$, respectively

for activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed. respectively (Figure 4.11). As direct evidence of nitrification, the experiments showed that more than 60 % of the influent alkalinity as calcium carbonate $CaCO_3$ was consumed with pH value, was between 7.21 ± 0.20 to 7.22 ± 0.20 .

Meanwhile, removal of the carbonaceous substances in terms of COD and SS, the experiments showed that all three filtrations media-based bed which used in the pilot-scale downflow BAFs (figure 4.2) have almost the same removal efficiency of COD there are no major differences, with removal efficiency has reached 91.89%, 91.60%, 90.51%, for tCOD, and 91.47%, 90.96%, 90.51%, for sCOD, respectively for the activated carbon-based filtration media bed, sand-based filtration media bed, and the ceramic particle-based filtration media bed. (Qiu, Zhang, Wang, & Du, 2010) [149], comparison between zeolite, ceramic particle, and carbonate media-based bed and the removal efficiency of COD have almost the same and different nitrification mode. When the conventional nitrification and the middle phase of denitrification (NO₂ reduction to N₂) are inhibited, the NH₄⁺ was eliminated along with the COD consumption (Chai, et al., 2019) [33]. For the suspended solids SS was observed that the SS elimination in the activated carbon-based filtration media bed and the sandbased filtration media bed appeared improved than ceramic particle-based filtration media bed. However, the experiments showed that all three-filtration media used in the pilot-scale downflow BAFs were able to reach the mean final effluent concentration of suspended solids SS less than 12 mg/L. While the effluent of SS consent concentration is commonly set at ≤35 mg L⁻¹ (Budge & Gorrie, 1996) [27]. Where the experiments showed that the mean influent concentration was 59.50±48.59 mg/L, while the mean final effluent concentration was 4.00±1.92 mg/L, 4.62±2.50 mg/L, and 11.62±4.62 mg/L, with removal efficiency has reached 94.24%, 93.34%, and 83.27% (Figure 4.7), respectively for the activated carbon-based filtration media bed, sand-based filtration media bed, and the ceramic particle-based filtration media bed. (Chang, Tran, & Park, 2009) [36], indicated that zeolite BAFs removed SS somewhat better than carbonate BAFs or ceramic particle BAFs, which may be because the zeolite possessed superior iron exchange and adsorption properties.

Two hypotheses exist about the elimination of NH₄⁺ -N. Heterotrophic nitrifiers developed distinct enzymes to catalyze the N elimination, which resulted in the elimination of ammonia when ClO₃⁻were supplemented. They were efficient for both nitrification and denitrification, and virtually no NO₂-N or NO₃-N were produced in treated wastewater solely with ammonia (Robertson & Kuenen, 1990) [159]. ClO₃⁻ inhibited membrane-bound nitrate reductase (Nar) function by blocking nitrate transporters (NarK), but had no effect on periplasmic nitrate reductase (Nap) (Rusmana & Nedwell, 2004) [169]. Nap is probable to eliminate NO₃⁻-N to NO₂⁻-N than membrane nitrate reductase as energy is not needed to translate NO₃⁻ and product NO₂⁻ through the membrane (Daims, Lucker, & Wagner, 2016; Lucker, Nowka, Rattei, Spieck, & Daims, 2013; Lucker, et al., 2010) [45], [115], [116]. Denitrification via Nap would result in Lowered NO₃⁻ during the full procedure when ClO₃⁻ was added.

4.3.3 Denitrification Properties of Different Filtration Media in BAFs

The main role of the three pilot-scale downflow BAFs in the denitrification process was to nitrify ammonium ions into nitrite and/or nitrate as well as the removals of residual pollutants. Figures (4.4, 4.8, 4.9, 4.10) show the mean influent and effluents concentrations of ammonium-N NH_4^+ -N, nitrite-N, NO_2^- -N, nitrate-N, NO_3^- -N, and total-KN (TKN).

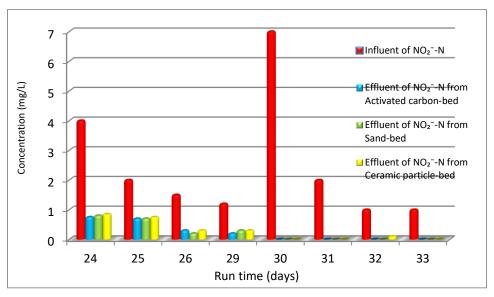


Figure 4.8. The influent and effluent concentration of NO₂-N comparison.

Evaluated the performance of pilot-scale downflow BAFs using activated carbon-based filtration media bed, via NO₂⁻-N or NO₃⁻-N, when the mean influent of NO₂⁻-N was 2.46 ± 2.07 mg/L (Figure 4.7), the mean sCOD concentration decreased from 41.50 ± 12.86 mg/L to 3.53 ± 0.79 mg/L, with a elimination efficacy of 91.47%. The mean effluent of NO₂⁻-N was 0.24 ± 0.31 mg/L, and the NO₃⁻-N level reduced from 1.99 ± 1.76 mg/L to 0.16 ± 0.24 mg/L (Figure 4.8), with a removal efficiency of 90.10%, 91.84%, respectively for NO₂⁻-N, and NO₃⁻-N. The mean mass removal of 0.19 ± 0.18 kg NO₂⁻-N/m³.d, and 0.15 ± 0.15 kg NO₃⁻-N/m³.d, via denitrification (figure 4.11).The mean total nitrogen TN concentration reduced from 32.12 ± 26.93 mg/L to 7.62 ± 4.49 mg/L (Figure 4.10), with a removal efficiency of 76.26%.

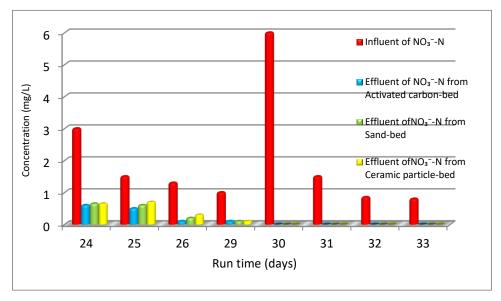


Figure 4.9. The influent and effluent concentration of NO₃-N comparison.

The evaluated the performance of pilot-scale downflow BAFs using sand-based filtration media bed, via NO₂⁻-N or NO₃⁻-N, when the mean influent of NO₂⁻-N was 2.46 ± 2.07 mg/L (Figure 4.7), the mean sCOD concentration decreased from 41.50 ± 12.86 mg/L to 3.75 ± 0.84 mg/L, with an elimination efficacy of 90.96%. The mean effluent of NO₂⁻-N was 0.25 ± 0.32 mg/L, and the NO₃⁻-N level reduced from 1.99 ± 1.76 mg/L to 0.19 ± 0.27 mg/L (Figure 4.8), with a removal efficiency of 89.84%, 90.28, respectively for NO₂⁻-N, and NO₃⁻-N. The mean mass removal of 0.19 ± 0.18 kg NO₂⁻-N/m³.d, via denitrification (figure 4.11). The mean TKN concentration reduced from 32.12 ± 26.93 mg/L to 7.93 ± 4.79 mg/L (Figure 4.9), with a removal efficiency of 75.29%.

Evaluated the performance of pilot-scale downflow BAFs using ceramic particle-based filtration media bed, via NO₂⁻-N or NO₃⁻-N, when the mean influent of NO₂⁻-N was 2.46 ± 2.07 mg/L (Figure 4.7), the mean sCOD concentration decreased from 41.50 ± 12.86 mg/L to 3.93 ± 0.97 mg/L, with an elimination efficacy of 90.51%. The mean effluent of NO₂⁻-N was 0.28 ± 0.34 mg/L, and the NO₃⁻-N level reduced from 1.99 ± 1.76 mg/L to 0.21 ± 0.29 mg/L (Figure 4.8), with a removal efficiency of 88.32%, 89.03%, respectively for NO₂⁻-N, and NO₃⁻-N. The mean mass removal of 0.18 ± 0.18 kg NO₂⁻-N/m³.d, and 0.15 ± 0.15 kg NO₃⁻-N/m³.d, via denitrification (figure 4.11). The mean TKN concentration reduced from 32.12 ± 26.93 mg/L to 8.08 ± 4.82 mg/L (Figure 4.10), with a removal efficiency of 74.82%.

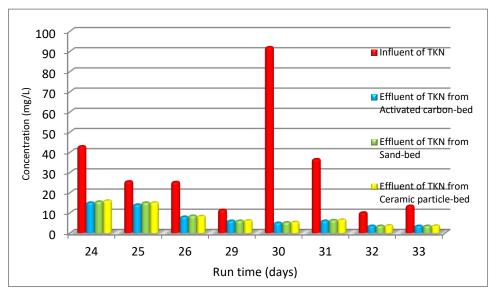


Figure 4.10. The influent and effluent concentration of TKN comparison.

(Hwang, Sakuma, & Tanaka, 1994) [88], conducted several batch experiments and pilot-scale trials on biological denitrification with submerged columns using isopropanol as the C-source. At an influent nitrate-N level of 20 mg/L and an isopropanol level of 40 mg/L, over 95% denitrification efficacy was detected in the pilot-scale analysis using submerged columns. (Ong, Hu, Lee, Ng, & Song, 2002 b)[133] and (Lee, Ong, & Ng, 2004)[134] demonstrated advancements in denitrification by the use of packed bed (PB) columns for high-rate N and C reduction. (Ong, Hu, Lee, Ng, & Song, 2002 b) [133], stated that the elimination rates of the anoxic-aerobic PB system were 7.41 and 28 kg COD/m³.day. The N and COD elimination efficacy of the anoxic-aerobic PB system was in the range of 97.5 - 100% and 98.6 - 99.4%, respectively. Furthermore, (Farabegoli, Gavasci, Lombard!, & Romani, 2003 b) [55] stated that the denitrification rate recorded 2.4 kg/m³.day for a water temperature of 25 °C in a tertiary up-flow sand filter. Moreover, an examination by (Lee, Ong, & Ng, 2004)[106] revealed that

the maximum possible TKN and COD elimination levels were 47.2 and 158.0 g COD/m².day and a dual-stage PB system was able to achieve TKN and COD elimination efficacy higher than 99 and 98%, respectively.

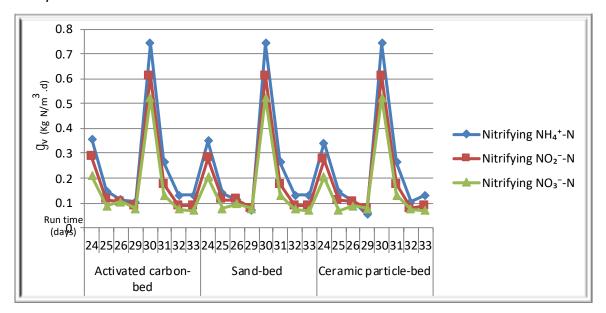


Figure 4.11. Mass removal of NH₄+-N, via nitrification NO₂--N, NO₃--N via denitrification comparison

4.4 Combined COD and Ammonium-Nitrogen Elimination – a Comparison Between Different Filtration Media on BAFs

The BAFs are three-stage reactors for wastewater treatment that use granular media for biomass support. BAFs are capable of achieving carbonaceous matter removal, solids filtration, nitrification, and/or denitrification in a single reactor. Three processes are used to eliminate carbonaceous contaminants from BAFs: solids filtration, absorption, and oxidation. Nitrification is a biological oxidation mechanism that requires two distinct bacterial communities. AOB performs the first step of nitrification by oxidizing ammonia to nitrite over hydroxylamine (NH₂OH); nitrite-oxidizing bacteria (NOB) complete the process by oxidizing nitrite to nitrate.

4.4.1 Start-up of the Experimental Trials

Throughout the duration of the experiments, all the three pilot-scale downflow BAFs were continuously operated fed with raw municipal wastewater containing various pollutants. All the three pilot-scale downflow BAFs were operated at ambient air temperature ranged between $8-29^{\circ}\text{C}$ with a mean of 17.93±7.27 °C. The mean influent flowrates were 0.18 L/min, and air: liquid ratio of 10: 1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and N loading rates of the three pilot-scales, downflow BAFs was $0.0022\pm0.00 \text{ kg NH}_4^+\text{-N/m}^3\text{.d}$, $0.0072\pm0.00 \text{ kg TKN/m}^3\text{.d}$, $0.0434\pm0.01\text{kg tCOD/m}^3\text{.d}$, $0.0285\pm0.00 \text{ kg sCOD/m}^3\text{.d}$, and $0.0931\pm0.17 \text{ kg SS/m}^3\text{.d}$.

4.4.2 Impact of Different Filtration Media on Reduction/ Removal of COD and Ammonium-Nitrogen

Figures 4.4, 4.12, and 4.13 show the mean influent and effluents concentrations of tCOD, sCOD, and NH_4^+ -N. Figure 4.14 showed the mean removal efficiency of tCOD, sCOD, and NH_4^+ -N comparison between three pilot-scale downflow BAFs with three different filtration media types demonstrated that all three media extremely well to the removal of tCOD, sCOD, and NH_4^+ -N.

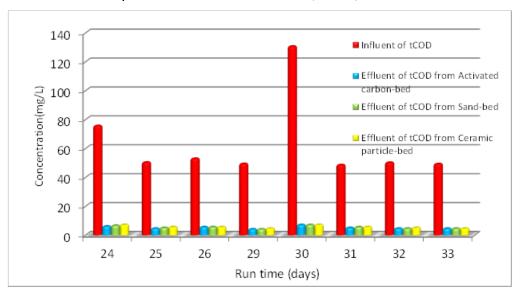


Figure 4.12. The influent and effluent concentration of tCOD comparison.

Performance of the pilot-scale downflow BAFs which used a 0.78 ± 0.60 mm activated carbon-based filtration media bed. Where the influent contains tCOD 63.25 ± 28.62 mg/L, sCOD 41.50 ± 12.86 mg/L, and NH₄⁺-N was 3.16 ± 2.46 mg/L, with recirculation of the influent 100%, at HRT 12-hours, Where the mean final effluent concentration of tCOD was 5.12 ± 0.99 mg/L, sCOD 3.53 ± 0.79 mg/L, and NH₄⁺-N was 0.31 ± 0.37 mg/L, with reduction/removal of 91.89%, 91.47%, and 90.11%, respectively for tCOD, sCOD, and NH₄⁺-N.

Performance of the pilot-scale downflow BAFs which used a 0.95 ± 0.58 mm sand-based filtration media bed. Where the influent contains tCOD 63.25 ±28.62 mg/L, sCOD 41.50 ±12.86 mg/L, and NH₄⁺-N was 3.16 ±2.46 mg/L, with recirculation of the influent 100%, at HRT 12-hours, Where the mean final effluent concentration of tCOD was 5.31 ± 1.03 mg/L, sCOD 3.75 ± 0.84 mg/L, and NH₄⁺-N was 0.38 ± 0.44 mg/L, with reduction/removal of 91.60%, 90.96%, and 87.74%, respectively for tCOD, sCOD, and NH₄⁺-N.

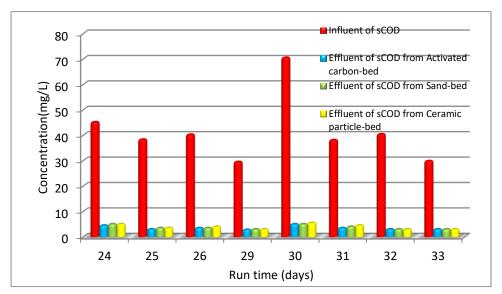


Figure 4.13. The influent and effluent concentration of sCOD comparison.

Performance of the pilot-scale downflow BAFs which used a 3.28 \pm 2.14 mm ceramic particle-based filtration media bed. Where the influent contains tCOD 63.25 \pm 28.62 mg/L, sCOD 41.50 \pm 12.86 mg/L, and NH₄⁺-N was 3.16 \pm 2.46 mg/L, with recirculation of the influent 100%, at HRT 12-hours, Where the mean final effluent concentration of tCOD was5.56 \pm 0.97 mg/L, sCOD 3.93 \pm 0.97 mg/L, and NH₄⁺-N was 0.46 \pm 0.46 mg/L, with reduction/removal of 90.51%, 90.51%, and 85.17%, respectively for tCOD, sCOD, and NH₄⁺-N.

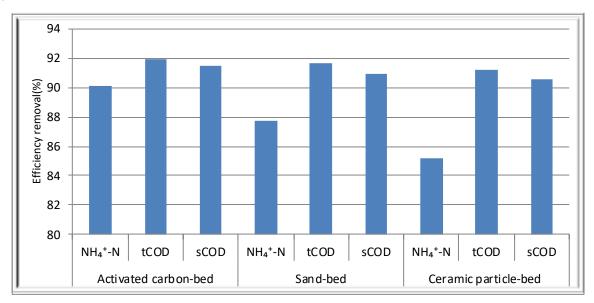


Figure 4.14. The mean efficiency removal of tCOD, sCOD, and NH₄+-N comparison.

The experimental results indicate that an activated carbon-based filtration media bed has a better capability for removal of ammonium-nitrogen NH₄⁺-N than a sand-based filtration media bed and ceramic particle-based filtration media bed in the nitrification process. However, all three media used in the pilot-scale downflow BAFs were able to reach the mean final effluent level of NH₄⁺-N less

than 0.47 mg/L. Meanwhile, removal of the COD the experiments showed that all three media-based bed have almost the same removal efficiency of COD there are no major differences, with a mean final effluent concentration of tCOD between 5.12 ± 0.99 mg/L to 5.56 ± 0.97 mg/L, and for sCOD between 3.53 ± 0.79 mg/L to 3.93 ± 0.97 mg/L.

According to (Gonzalez-Martinez & Wilderer, 1991) [66], in the downflow of BAFs where carbonaceous matter and ammonia are removed simultaneously, microorganisms living in the filter's low region receive rich-oxygen-air rich and hence would not suffer oxygen inadequacy. (Peladan, Lemmel, & Pujol, 1996; Tschui, Boller, Gujer, Eugster, Mader, & Stengel, 1994; Husovitz, 1998)[139], [197], [86], found a modest increase in nitrification efficiency when water velocity was increased through recirculation, which contributed to increased ammonia mass transfer into the biofilms. Another possibility is that the recirculation of the effluent dilutes the influent ammonia and sCOD, causing the lowered level of ammonia that can be converted over a shorter hydraulic holding period (Ha, Ong, & Surampalli, 2010) [69].

4.4.3 Influence of COD and Ammonium-Nitrogen Loading of the Influent on the Nitrification at Different Filtration Media

For the pilot-scale downflow, BAFs used a 0.78 ± 0.60 mm activated carbon-based filtration media bed. When the mean influent organic N loading rates was 0.0022 ± 0.00 kg NH₄⁺-N/m³.d, 0.0072 ± 0.00 kg TKN/m³.d, 0.0434 ± 0.01 kg tCOD/m³.d, 0.0285 ± 0.00 kg sCOD/m³.d, and 0.0931 ± 0.17 kg SS/m³.d, with recirculation of the influent 100%, at HRT 12-hours, the activated carbon-based filtration media bed was able to nitrify 0.24 ± 0.21 kg NH₄⁺-N/m³.d, and 0.78 ± 0.54 kg TKN/m³.d. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS. Where the mean mass removal was 5.07 ± 2.42 kg tCOD/m³.d, 3.31 ± 1.06 kg sCOD/m³.d, and 5.72 ± 4.08 kg SS/m³.d.

For the pilot-scale downflow BAFs which used a 0.95 ± 0.58 mm sand-based filtration media bed. When the mean influent organic and nitrogen loading rates was 0.0022 ± 0.00 kg $NH_4^+-N/m^3.d$, 0.0072 ± 0.00 kg $TKN/m^3.d$, 0.0434 ± 0.01 kg $tCOD/m^3.d$, 0.0285 ± 0.00 kg $tCOD/m^3.d$, and 0.0931 ± 0.17 kg $tCOD/m^3.d$, with recirculation of the influent $tCOD/m^3.d$, at HRT $tCOD/m^3.d$, the sand-based filtration media bed was able to nitrify $tCOD/m^3.d$, and $tCOD/m^3.d$, and $tCOD/m^3.d$. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in terms of $tCOD/m^3.d$, and $tCOD/m^3.d$, and an achieved and achieved an achieved an

For the pilot-scale downflow, BAFs used a 3.28 ± 2.14 mm ceramic particle-based filtration media bed. When the mean influent organic and nitrogen loading rates was 0.0022 ± 0.00 kg NH₄+-N/m³.d, 0.0072 ± 0.00 kg TKN/m³.d, 0.0434 ± 0.01 kg tCOD/m³.d, 0.0285 ± 0.00 kg sCOD/m³.d, and 0.0931 ± 0.17 kg SS/m³.d, with recirculation of the influent 100%, at HRT 12-hours, the ceramic particle-based filtration media bed was able to nitrify 0.23 ± 0.22 kg NH₄+-N/m³.d, 0.76 ± 0.54 kg TKN/m³.d. Meanwhile, was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS. When the mean mass removal was 5.03 ± 2.43 kg tCOD/m³.d, 3.28 ± 1.05 kg sCOD/m³.d, and 5.05 ± 3.85 kg SS/m³.d.

The results of this study showed that all three filtration media used in the pilot-scale downflow BAFs was able to nitrify between 0.23 ± 0.22 kg NH₄⁺-N/m³.d to 0.24 ± 0.22 kg NH₄⁺-N/m³.d, and between 0.76 ± 0.54 kg TKN/m³.d to 0.78 ± 0.54 kg TKN/m³.d. (Ha, Ong, & Surampalli, 2010) [69], reported that

the ammonia mass eliminated for the 3-media were statistically different; for the gravel BAFs, The mass of ammonia extracted increased to about 0.5 kg NH₃-N/m³.d. and remained relatively stable as the ammonia loading was increased further. (Rogalla & Sibony, 1992) [163], stated that nitrification rates were about 0.6 kg N/m³.d. (Peladan, Lemmel, & Pujol, 1996) [139], performed a pilot analysis with the Biofor method for water velocities ranging from 5-20 m h⁻¹. The pilot plant was capable to nitrify 2.7 kg NH₃-N/m³.d at 14 °C. For reducing/elimination of the carbonaceous, study results indicated that all three filtration media used in the pilot-scale downflow BAFs were able to reduce/remove between 5.03±2.43 kg tCOD/m³.d to 5.07±2.42 kg tCOD/m³.d, and 3.28±1.05 kg sCOD/m³.d to 3.31±1.06 kg sCOD/m³.d. For the suspended solids were able to reduce/eliminate between 5.05±3.85 kg SS/m³.d to 5.72±4.08 kg SS/m³.d. According to (Rogalla, Lacamp, Bacquet, & Hansen) [164], in BAFs with fixed bed and dense media can treat volumetric loads, around 5 - 10 kg COD/m³.d.

4.5 The ratio sCOD/TKN - Influence on the Nitrification Process and Comparison Between Filtration Media in BAFs

The stoichiometry sCOD/TKN ratio describes the amount of organic carbon required to remove nitrate from the wastewater. It is essential to study the impact of the sCOD/TKN ratio on nitrogen elimination from wastewater. It should be remembered, nevertheless, that pre-treatment of raw wastewater for SS removal can result in the loss of organic matter, lowering the (sCOD/TKN ratio) of the wastewater. An adequate sCOD/TKN ratio must be supplemented to fully denitrify the nitrate produced during the N elimination process.

4.5.1 Start-up of the Experimental Trials

Three pilot-scale downflow BAFs were operated at ambient air temperature ranged from 8 to 29° C with a mean of 17.93±7.27 °C. The mean influent flowrates were 0.18 L/min, and air: liquid ratio of 10:1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100%,these experiments with three different sCOD/TKN ratios (2.8, 4.0, 4.5). Run 30 sCOD/TKN ratio = 2.8 where a feed carbon and nitrogen loading was 0.0483 Kg sCOD/m³.d, 0.0895 kg tCOD/m³.d, and 0.0058 kg NH₄+-N/m³.d. Run 25 sCOD/TKN ratio = 4.0 where the loading was 0.0262 Kg sCOD/m³.d, 0.0344 kg tCOD/m³.d, and 0.0017 kg NH₄+-N/m³.d. Run 29 sCOD/TKN ratio = 4.5 where the loading was 0.0202 Kg sCOD/m³.d, 0.0337 kg tCOD/m³.d, and 0.0011 kg NH₄+-N/m³.d.

4.5.2 Effects of sCOD/TKN ratio on Nitrogen Removal at Different Filtration Media

Run time 30 (sCOD/TKN ratio of 2.8), Summary data of experiments during the monitoring period., and pH between 7.30 to 7.40. The recirculation of the influent 100% at HRT 12-hours, were the mean influent concentration of NH_4^+ -N 8.50 mg/L, NO_2^- -N 7.00 mg/L, and NO_3^- -N 6.00 mg/L. As results of the experiments show that removal of 100% of the NH_4^+ -N, NO_2^- -N, and NO_3^- -N at activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed (Figures 4.15, 4.16, and 4.17). All the three filtrations media-based bed using in the three pilot-scale downflow BAFs was able to nitrify 0.7425 kg NH_4^+ - N/m^3 .d, 0.6114 kg NO_2^- - N/m^3 .d, and 0.5241 kg NO_3^- - N/m^3 .d, respectively for each pilot-scale reactor. There are two possible explanations for the elimination of NH_4^+ -N. Heterotrophic nitrifiers provided distinct enzymes to catalyze the nitrogen removal operation, while ammonia elimination takes place in the presence of ATU and ClO₃.

They were essential for nitrification and denitrification, and virtually no NO₂-N or NO₃-N was collected as wastewater was treated solely with ammonia (Robertson & Kuenen, 1990) [159]. ClO₃ could inhibit membrane-bound nitrate reductase (Nar) action by inhibiting nitrate transporters (NarK), but it could not hinder periplasmic nitrate reductase (Nap) (Rusmana & Nedwell, 2004) [169]. Nap is possible to eliminate NO₃⁻-N to NO₂⁻-N than membrane nitrate reductase as energy is not needed to translate NO₃⁻ and product NO₂⁻through the membrane (Daims, Lucker, & Wagner, 2016; Lucker, Nowka, Rattei, Spieck, & Daims, 2013; Lucker, et al., 2010) [45], [115], [116]. The denitrification via Nap would result in lowering NO₃⁻ level during the whole procedure when adding ClO₃⁻.

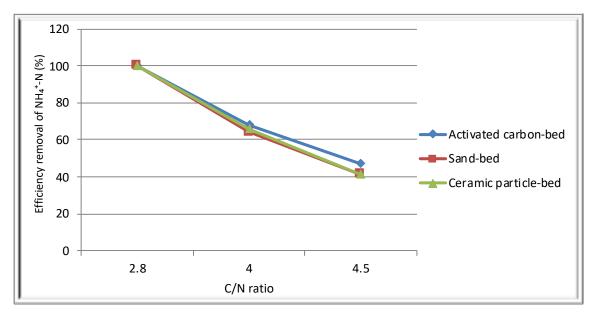


Figure 4.15. The efficiency removal of NH₄+-N comparison.

Run time 25 (sCOD/TKN ratio of 4.0), during these experiments the DO levels were preserved between 1.98 mg/L to 2.05 mg/L, and pH around 7.0. Recirculation of the influent 100% at HRT 12-hours, were the mean influent concentration of NH₄⁺-N 2.50 mg/L, NO₂⁻-N 2.00 mg/L, and NO₃⁻-N 1.50 mg/L. As a result of the experiments show that activated carbon-based filtration media bed achieved efficiency removal of 68.00%, 65.00%, and 66.66% (Figures 4.15, 4.16, and 4.17), where the mean final effluent level was 0.80 mg/L, 0.70 mg/L, and 0.50 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.1485 kg NH₄⁺-N/m³.d, 0.1135 kg NO₂⁻-N/m³.d, and 0.0873 kg NO₃⁻-N/m³.d. Furthermore, the sand-based filtration media bed achieved efficiency removal of 64.0%, 65.0%, and 60.0%, where the mean final effluent level was 0.90 mg/L, 0.70 mg/L, and 0.60 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.1397 kg NH₄⁺-N/m³.d, 0.1135 kg NO₂⁻-N/m³.d, and 0.0786 kg NO₃⁻-N/m³.d. Meanwhile, the ceramic particle-based filtration media bed achieved efficiency removal of 66.0%, 62.5%, and 53.3%, where the mean final effluent level was 0.85 mg/L, 0.75 mg/L, and 0.70 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.1441 kg NH₄⁺-N/m³.d, 0.1091kg NO₂⁻-N/m³.d, and 0.0698 kg NO₃⁻-N/m³.d.

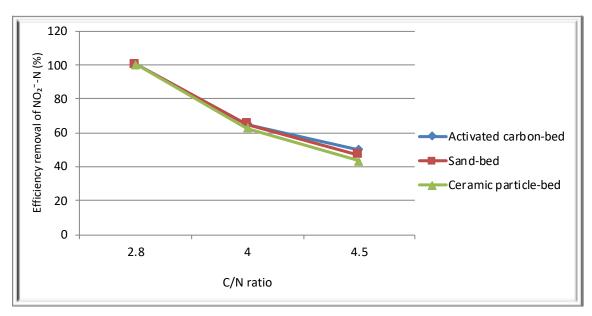


Figure 4.16. The efficiency removal of NO₂⁻-N comparison.

Run time 29 (sCOD/TKN ratio of 4.5), during these experimental the DO levels were kept DO between 3.5 mg/L to 4.15 mg/L, and pH between 7.00 to 7.30. The recirculation of the influent 100% at HRT 12-hours, was the mean influent level of NH₄⁺-N 1.70 mg/L, NO₂⁻-N 1.50 mg/L, and NO₃⁻-N 1.30 mg/L. A result of the experiments show that activated carbon-based filtration media bed achieved elimination efficiency of 47.05%, 50.00%, and 53.84% (Figures 4.15, 4.16, and 4.17), where the mean final effluent level was 0.90 mg/L, 0.75 mg/L, and 0.60 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.1048 kg NH₄⁺-N/m³.d, 0.0873 kg NO₂⁻-N/m³.d, and 0.0786 kg NO₃⁻-N/m³.d. Furthermore, the sand-based filtration media bed achieved elimination efficacy of 41.17%, 46.66%, and 50.00%, where the mean final effluent level was 1.00 mg/L, 0.80 mg/L, and 0.65 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.0698 kg NH₄⁺-N/m³.d. Meanwhile, the ceramic particle-based filtration media bed achieved efficiency removal of 41.17%, 43.33%, and 50.00%, where the mean final effluent concentration was 1.00 mg/L, 0.85 mg/L, and 0.65 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N, in addition, was able to nitrify 0.0524 kg NH₄⁺-N/m³.d, 0.0786 kg NO₂⁻-N/m³.d, and 0.0786 kg NO₃⁻-N/m³.d.

The results of this study showed that all three filtration media used in the pilot-scale downflow BAFs with three different sCOD/TKN ratios (2.8, 4.0, 4.5) indicate that as the C/N ratio increased, the nitrogen (NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N) elimination (%) decreased linearly (Figures 4.15, 4.16 and 4.17), as well as, the C/N ratio increased, the mass of the nitrogen removed (NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N) per unit volume (kg/m³.d) decreased linearly. Correspondingly, the effluent of the nitrogen (NH₄⁺-N, NO₂⁻-N, and NO₃⁻-N) concentrations increased as the COD/TKN ratio increased. The results of this study showed that all three-filtration media used in the pilot-scale downflow BAFs was extremely well in the nitrogen conversion and removal. However, of the three-filtration media-based bed types, the pilot-scale downflow BAFs which used a 0.78±0.60 mm activated carbon-based filtration media bed showed the highest percentage of nitrogen elimination efficacy and mass the N

eliminated at three different COD/TKN ratios. (Ha J. , 2006) [70], stated that various C/N ratios are expressed as the C/N ratio increased, the ammonia elimination (%) and ammonia mass eliminated lowered linearly. Respectively, the effluent ammonia concentrations increased as the C/N ratio increased. However, a different study, (Ballinger, Head, Curtis, & Godley, 2002) [15], revealed that the influence of influent C/N ratio (2-to-5) on the β -proteobacterial autotrophic AOB structure by DGGE evaluation of 16S rRNA gene fragments amplified with a range of AOB-selective primers. At a C/N ratio of 2, nitrification was estimated around 1.0 mg NH₄+-N/g-DW of biomass/h and at a C/N ratio of 5, there was a 50% decrease in nitrification levels. Quantitative FISH analysis revealed that β -proteobacterial AOB is present at a concentration of approximately 108 cells/mL of biomass with a C/N ratio of 2, but was not observed at a C/N ratio of 5.

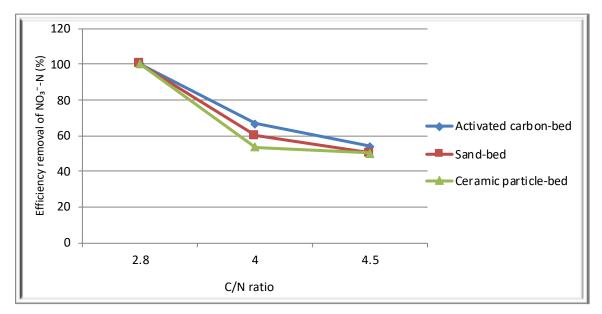


Figure 4.17. The efficiency removal of NO₃⁻-N comparison.

4.5.3 Effects of sCOD/TKN ratio on COD reduction/removal at three different filtration media types

Carbonaceous, in terms of COD, is a problem in wastewater treatment. As a result, a wastewater treatment facility's primary objective is to remove it. The microorganisms that grow attached to the filter packed material carried out the removal process. In the current study at Run time 30 (C/N ratio of 2.8), during this experiment maintained DO between 6.00 mg/L to 6.10 mg/L, and pH between 7.30 to 7.40. The recirculation of the influent 100% at HRT 12-hours, was the mean influent concentration of the sCOD 70.50 mg/L, and the tCOD 130.50 mg/L. As a result of the experiments show that the pilot-scale downflow BAFs using activated carbon-based filtration media bed achieved removal efficiency was high at sCOD 92.90%, and the tCOD was 94.63% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 5.00 mg/L, and the tCOD 7.00 mg/L. In addition the mean mass removal was 5.7216 kg sCOD/m³.d, and 10.7881 kg tCOD/m³.d. Meanwhile, the pilotscale downflow BAFs using a sand-based filtration media bed achieved removal efficiency was high at sCOD 92.90%, and the tCOD 94.63% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 5.00 mg/L, and the tCOD 7.00 mg/L. In addition the mean mass removal was 5.7216 kg sCOD/m³.d, and 10.7881 kg tCOD/m³.d. Likewise, the pilot-scale downflow BAFs using a ceramic particle-based filtration media bed achieved removal efficiency was high at sCOD 92.19%, and the tCOD 94.63% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 5.50 mg/L, and the tCOD 7.00 mg/L. in addition the mean mass removal was 5.6779kg sCOD/m³.d, and 10.7881 kg tCOD/m³.d.

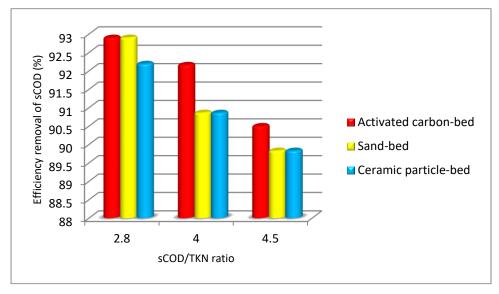


Figure 4.18. The efficiency removal of sCOD comparison.

Run time 25 (sCOD/TKN ratio of 4.0), during this experiment the DO levels were maintained between 1.98 mg/L to 2.05 mg/L, and pH around 7.00. The recirculation of the influent 100% at HRT 12-hours, was the mean influent concentration of the sCOD 38.30 mg/L, and the tCOD 50.25 mg/L. As a result of the experiments show that the pilot-scale downflow BAFs using activated carbon-based filtration media bed achieved removal efficiency was high at sCOD 92.16%, and the tCOD 91.04% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 3.00 mg/L, and the tCOD 4.50 mg/L, in addition, the mean mass removal was 3.0835 kg sCOD/m³.d, and 3.9964 kg tCOD/m³.d. Meanwhile, the pilot-scale downflow BAFs using a sand-based filtration media bed achieved removal efficiency was high at sCOD 90.86%, and the tCOD 90.04% (Figures 4.18, and 4.19), with the mean mass removal was 3.0398 kg sCOD/m³.d, and 3.9527 kg tCOD/m³.d. Likewise, the pilot-scale downflow BAFs using a ceramic particle-based filtration media bed achieved removal efficiency was high at sCOD 90.86%, and the tCOD 89.05% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 3.50 mg/L, and the tCOD 5.50 mg/L. in addition the mean mass removal was 3.0398 kg sCOD/m³.d, and 3.9990 kg tCOD/m³.d.

Run time 29 (sCOD/TKN ratio of 4.5), during this experiment the DO levels were maintained between 3.5 mg/L to 4.15 mg/L, and pH between 7.00 to 7.30. The recirculation of the influent 100% at HRT 12-hours, was the mean influent concentration of the sCOD 29.50 mg/L, and the tCOD 49.25 mg/L. As a result of the experiments show that the pilot-scale downflow BAFs using activated carbon-based filtration media bed achieved removal efficiency was high at sCOD 90.50%, and the tCOD 91.87% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 2.80 mg/L, and the tCOD 4.00 mg/L, in addition, the mean mass removal was 2.3323 kg sCOD/m³.d, and 3.9527 kg tCOD/m³.d. Meanwhile, the pilot-scale downflow BAFs using a sand-based filtration media bed achieved removal efficiency was at sCOD 89.83%, and the tCOD 91.87% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 3.00 mg/L, and the tCOD 4.00 mg/L, in addition, the mean mass removal was 2.3148 kg sCOD/m³.d, and 3.9527 kg tCOD/m³.d. Likewise, the pilot-

scale downflow BAFs using a ceramic particle-based filtration media bed achieved removal efficiency was at sCOD 89.83%, and the tCOD 90.86% (Figures 4.18, and 4.19), with the mean final effluent concentration was at sCOD 3.00 mg/L, and the tCOD 4.50 mg/L. in addition, the mean mass removal was 2.3148 kg sCOD/m³.d, and 3.9090 kg tCOD/m³.d.

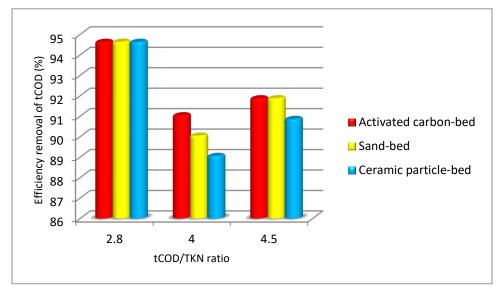


Figure 4.19. The efficiency removal of tCOD comparison. (Activated carbon-bed, Sand-bed, Ceramic particle-bed)

The experiments with three different COD loading and three different sCOD/TKN ratios (2.8, 4.0, 4.5). The experimental results indicate that as the C/N ratio increased, the COD (sCOD, and tCOD) loading, and COD (sCOD, and tCOD) removal (%) decreased (Figures 4.18, and 4.19), Correspondingly, the C/N ratio decreased, the mass of the COD removed (sCOD, and tCOD) per unit volume (kg/m³.d), and removal (%) increased. A potential explanation is that heterotrophs and nitrifiers compete for oxygen uptake, and nitrifiers within the biofilm are affected by heterotrophs' accelerated growth in the presence of increased COD, which could reduce the oxygen interaction level of ammonium oxidizing bacteria or nitrifying bacteria (Satoh, Okabe, Norimatsu, & Watanabe, 2000). In another study (Fatihah, 2004), reported that for different C/N ratios, the COD elimination efficacy did not significantly differ with the ratio.

4.6 Effect of DO, Alkalinity, and pH on Nitrification by DifferentFiltration Media in BAFs

DO levels have a strong impact on nitrifying bacteria' growth rates. Incomplete nitrification resulted in the presence of low DO, resulting in an ammonium build-up within the BAFs due to inadequate aeration time to transform the ammonia to nitrate. Meanwhile, alkalinity is critical not only for nitrification but also for system stabilization, since a reduction in pH is caused by the elimination of ammonium-nitrogen NH₄⁺-N from the system, and ammonium was also found to be closely associated with wastewater alkalinity.

4.6.1 Start-up of the Experimental Trials

Throughout the duration of the experiments, all the three pilot-scale downflow BAFs were continuously operated fed with raw municipal wastewater. All the three pilot-scale downflow BAFs were operated at ambient air temperature ranged from 8 °C to 29°C with a mean of 17.93 \pm 7.27 °C. The mean influent flowrates were 0.18 L/min, and the air: liquid ratio of 10: 1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and nitrogen loading rates of the three pilot-scale downflow BAFs was 0.0022 \pm 0.00 kg NH₄+-N/m³.d, 0.0072 \pm 0.00 kg TKN/m³.d, 0.0434 \pm 0.01kg tCOD/m³.d, 0.0285 \pm 0.00 kg sCOD/m³.d, and 0.0931 \pm 0.17 kg SS/m³.d.

4.6.2 Effect of DO Concentration on Nitrification

Dissolved oxygen has a strong impact on nitrifying bacteria's growth rates. Incomplete nitrification resulted in low DO, resulting in an ammonium build-up within the BAFs (owing to a lack of aeration time for the ammonia to be converted to nitrate). (Zhu & Chen, 2002), stated that another significant factor in maintaining DO in the fixed film processes was to keep ample DHT on hand due to the high diffusion of DFT in those operations.

As results of the experimental trials show that in the pilot-scale downflow BAFs using activated carbon-based filtration media bed when the mean concentration of dissolved oxygen was 6.76 ± 2.25 mg/L the mean nitrification efficiency has reached 90.11%. In the pilot-scale downflow BAFs using a sand-based filtration media bed, when the mean concentration of dissolved oxygen was 7.33 ± 1.82 mg/L the mean nitrification efficiency has reached 87.74 %. In the pilot-scale downflow BAFs using a ceramic particle-based filtration media bed, when the mean concentration of dissolved oxygen was 6.37 ± 1.84 mg/L, the mean nitrification efficiency has reached 85.17%. relative to dissolved oxygen to the percentage nitrification was plotted as shown in figure 4.20.

The DO half-saturation coefficients of Nitroso-bacteria or Ammonia Oxidizing Bacteria AOB and Nitro-bacteria or Nitrite oxidizer Bacteria NOB are 0.2 - 0.4 mg/L and 1.2 - 1.5 mg/L, respectively (Picioreanu, van Loosdrecht, & Heijnen, 1997) [144]. Thus, no overlying biomass restricts the growth of the NOB, which increases the levels of nitrite in the water (Peng Y. , et al., 2004) [142]. While numerous researchers suggested that decreased DO may impede NOB growth and result in AOB accumulation, the essential DO values published in the literature varied. (Laanbrock & Gerards, 1993; Wyffels, et al., 2004; Peng & Zhu, 2006) [103], [220], [140].

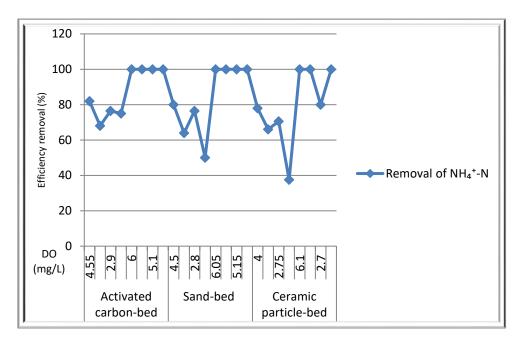


Figure 4.20. Relationship between DO and NH₄⁺-N removal efficiency comparison.

The amount of O_2 that should be transported in the aeration tank is equal to the amount of O_2 needed to oxidise the organic materials by the microorganisms in the activated sludge environment. DO amounts have a strong effect on nitrifying bacterial growth rates. When conducting nitrification, DO levels are usually retained at or above 2 mg/L in the reactor (because the growth rate of nitrifying bacteria slows significantly below 2 mg/L). According to Zhu and Chen, it was more critical to sustain ample DO in the fixed film procedures than in the suspended growth process due to the fixed film's nature of diffusion transport (Zhu & Chen, 2002) [227]. Additionally, the nitrification rate increases as dissolved oxygen concentration increases, and it is observed to be almost linearly correlated to oxygen concentrations greater than 10 mg/L. (Kermani, Bina, Movahedian, Amin, & Nikaeen, 2009) [99]. However, values greater than 4 mg/L do not substantially enhance activities, since it has been shown that excessive aeration results in decreased nitrification performance due to the detachment of the biofilm from the plastic media. (Peng Y., et al., 2004) [142]. Consequently, the nitrification rate increases with rising DO concentrations in the range of 1-3 mg/L and decreases with increasing concentrations in the range of 0.3-0.5 mg/L (Qasim, 1998) [148]. If the issue is caused by a lack of oxygen, simultaneous nitrification and denitrification (SND) can occur instead, which is normally checked by running the aeration equipment at maximum capacity or by lowering the device SRT, if necessary, to reduce the oxygen demand. As a result, the aeration equipment should have sufficient ability to sustain a dissolved oxygen concentration of at least 2.5 mg/L in the aeration tank under standard loading conditions. If 2.5 mg/L of oxygen cannot be sustained, it could be necessary to upgrade the current aeration device.

4.6.3 Effect of Alkalinity and pH on Nitrification

Alkalinity is also an estimate of the amount of water alkalinity as well as an indicator of the system's overall stability. The reduction in pH is attributable to the elimination of NH₄-nitrogen-ammonium from the environment, and the degree of alkalinity in the wastewater has increased as a result.

Figure 4.21 depicts the relationship between alkalinity (CaCO₃) and ammonium (NH₄) removal efficiency as CaCO₃ concentration increases. The result of these experimental trials shows that the mean influent alkalinity was 207.25 ± 8.97 mg CaCO₃/L during the entire period of the experimental trials. As direct evidence of nitrification, more than 60 % of the influent alkalinity as CaCO₃ was consumed. While the mean effluent of NH₄⁺-N oxidized was 0.31 ± 0.37 , 0.38 ± 0.44 , and 0.46 ± 0.46 mg NH₄⁺-N/L, respectively for the activated carbon-based filtration media bed, sand-based filtration media bed, and ceramic particle-based filtration media bed. In order to achieve full nitrification, adequate alkalinity must be present. With an improvement in alkalinity, the alkalinity impacting nitrification and the nitrogen removal rate increased (Sakairi, Yasuda, & Matsumura, 1996) [176].

A result of these experimental trials shows that in the pilot-scale downflow BAFs using activated carbon-based filtration media bed, the mean effluent alkalinity was 77.25 ± 2.60 mg/ L CaCO₃ when the mean pH was 7.21 ± 0.20 the mean nitrification efficiency has reached 90.11%. In the pilot-scale downflow BAFs using a sand-based filtration media bed, the mean effluent alkalinity was 77.56 ± 2.77 mg/ L CaCO₃ when the mean pH was 7.22 ± 0.20 the mean nitrification efficiency has reached 87.74%. In the pilot-scale downflow BAFs using a ceramic particle-based filtration media bed, the mean effluent alkalinity was 77.31 ± 2.71 mg/ L CaCO₃ when the mean pH was 7.22 ± 0.20 the mean nitrification efficiency has reached 85.17% during the entire period of this study. The relationship between alkalinity and NH_4^+ -N removal efficiency is shown in figure 4.21, and the relationship between pH and NH_4^+ -N removal efficiency is shown in figure 4.22. However, the result of these experimental trials consistent with the result of (Gujer & Boller, 1986) [68], were stated that in nitrifying bio-filters applied in the treatment of municipal wastewater, alkalinity of at least 75 mg L⁻¹ was required to sustain the highest nitrification rate.

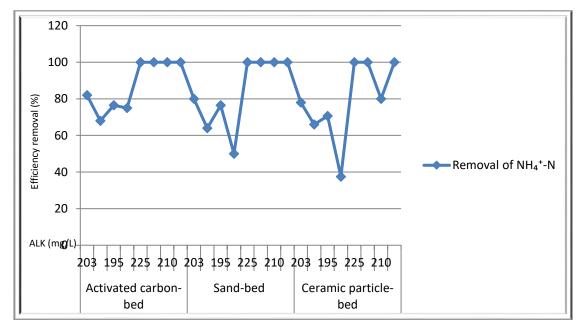


Figure 4.21. Relationship between alkalinity and NH₄⁺-N removal efficiency comparison.

The impact of alkalinity on nitrification rate is pH dependent; it has been suggested that the pH of wastewater between 6 and 9 protects species (Akpor, Momba, & Okonkwo, 2008) [6]. According to research on the impact of pH on nitrification efficacy in an up-flow biofilter, nitrification efficiency

increased linearly by 13% per unit pH rise from pH 5.0 to 8.5. (Villaverde, Garcia-Encina, & Fdz-Polanco, 1997 b) [211].

Alkalinity level is another significant wastewater feature that influences the biological nitrification process's efficiency. Appropriate alkalinity is required for completing nitrification. Additionally, alkalinity provides the buffering capability required to avoid pH changes caused by nitrification acid generation. Accordingly, the fact that the effect of alkalinity on the nitrification rate is often similar to the effect of pH, it can be shown that the rate of N elimination rises as alkalinity upsurges.

The NH₄⁺ was eliminated with the COD use when the conventional nitrification (Chai, et al., 2019) [33]. The pH value declines during the aeration process as a result of nitrification's intake of alkalinity. In theory, the pH value does not change during the conversion of nitrite to nitrate since no hydrogen ion is formed, as occurs during the conversion of ammonia to nitrite. However, if both nitrification and denitrification are complete, the pH can rise due to CO₂ stripping. As a result, there will be a change in the pH of the liquid form at the end of nitrification (Peng, Gao, Wang, & Sui, 2003) [143].

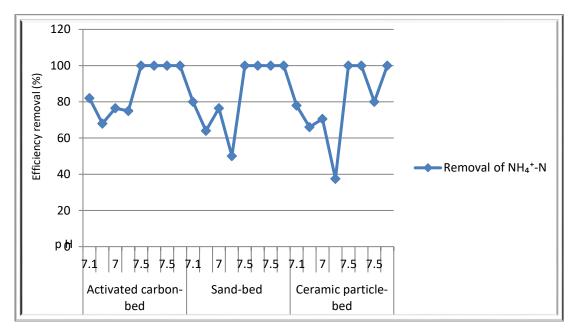


Figure 4.21. Relationship between pH and NH₄⁺-N removal efficiency comparison.

5 General Conclusions

5.1 General

In this chapter, the summary, overall conclusions, and the overall achievement of current research are expressed. Furthermore, suggested areas of the research that require further development derived from this thesis are also highlighted.

5.2 Structure of the Thesis

The thesis composed of 5 chapters, including this chapter which contains the background of the study, this allows defining the problem statement of the research, scope of research, the aim, and various goals consequent from this main objective as follows:

- Chapter 1. Background and Objectives It provides a review of Nitrogen Cycle, toxicity and
 effects on human body and a brief description of the wastewater treatment technologies for
 removal.
- Chapter 2. Nitrogen Removal Current Status It provides a review of current statusnitrogen elimination and technology of biological aerated filter systems BAFs including, possible removal mechanisms of nitrogen from wastewater in BAFs, and discussion of nitrification and denitrification principles.
- Chapter 3. Materials and methods It presents the experimental set-up and sample analyses, besides the nitrogen compound, and carbonaceous is also explained. Though, the detailed materials and methods applied, these experiments by three similar pilot-scale reactors of BAFs, a down-flow mode using activated carbon-based filtration media bed, sandbased filtration media bed, and ceramic particle-based filtration media bed as biomass support as an associated growth zone in wastewater treatment.
- Chapter 4. Results and discussion It presents the study outcomes & discussion of
 experiments during the monitoring period provided a comparison between three different
 filtration media types used as attached growth zones in the municipal wastewater treatment,
 for nitrogen conversion and removal by biological aerated filtration.
- Chapter 5. General conclusions and recommendations In this chapter, the main conclusions, and the key achievement of the current study are summarized. Moreover, suggested areas of the research that require further development resulting from the current thesis are also illustrated.
- References In this chapter, detailed references used in this thesis are presented.

5.3 Summary

The aim of this research was to examine the efficiency of nitrogen conversion and removal by biological aerated filtration BAFs. The laboratory experiments by three identical pilot-scale downflow of BAFs using three different filtration media types, 0.78 ± 0.60 mm activated carbon-based filtration media bed, 0.95 ± 0.58 mm sand-based filtration media bed, and 3.28 ± 2.14 mm ceramic particle-based filtration media bed as biomass support as attached growth zones in the treatment of municipal wastewater to the development of local materials bed, low cost, for improving the effluent quality before discharge to the aquatic environment.

Throughout the duration of the experiments, all the three pilot-scale downflow BAFs were continuously operated fed with raw municipal wastewater containing various pollutants, e.g. ammonium-nitrogen, nitrite-nitrogen, and nitrate-nitrogen. All the three pilot-scale downflow BAFs were operated at ambient air temperature ranged from 8 °C to 29°C with a mean of 17.93 \pm 7.27 °C. The mean influent flowrates were 0.18 L/min, and the air: liquid ratio of 10: 1. The hydraulic retention time HRT was 12-hours. The recirculation of the influent was 100% with daily backwashes. As a feed organic and nitrogen loading rates of the three pilot-scale downflow BAFs was 0.0022 \pm 0.00 kg NH₄+-N/m³.d, 0.0072 \pm 0.00 kg TKN/m³.d, 0.0434 \pm 0.01kg tCOD/m³.d, 0.0285 \pm 0.00 kg sCOD/m³.d, and 0.0931 \pm 0.17 kg SS/m³.d.

The efficiency and nitrification properties of three pilot-scale downflow BAFs with an activated carbon-based filtration, sand-based filtration, and ceramic particle-based filtration, respectively, have been examined to assess the practicability of applying studied media as biomass support as attached growth zone in BAFs. The nitrification and denitrification were first observed on day 24 where all the three pilot-scale downflow BAFs have achieved steady-state conditions, where the number of microorganisms increased, nitrification dominated. All the three-filtration media-based bed using in the three pilot-scale downflow BAFs was extremely well in the nitrogen conversion and removal, as showing in Table 5.1.

Table 5.1. Summary data of experiments during the monitoring period.

Parameter	Municipal wastewater	BAFs use	e downflow d activated based bed	Pilot-scale downflow BAFs used sand-based material bed		Pilot-scale downflow BAFs used ceramic particle-based bed	
	Influent (mg/L)	Effluent (mg/L)	Removal efficiency (%)	Effluent (mg/L)	Removal efficiency (%)	Effluent (mg/L)	Removal efficiency (%)
NH ₄ +-N	3.16±2.46	0.31±0.37	90.118	0.38±0.44	87.747	0.46±0.46	85.179
NO ₂ -N	2.46±2.07	0.24±0.31	90.103	0.25±0.32	89.847	0.28±0.34	88.324
NO ₃ -N	1.99±1.76	0.16±0.24	91.849	0.19±0.27	90.284	0.21±0.29	89.030
TKN	10.46±6.74	1.42±0.95	86.379	1.58±0.97	84.826	1.70±1.00	83.692
tCOD	63.25±28.62	5.12±0.99	91.898	5.31±1.03	91.602	5.56±0.97	90.514
sCOD	41.50±12.86	3.53±0.79	91.478	3.75±0.84	90.965	3.93±0.97	90.514
SS	59.50±48.59	4.00±1.92	94.244	4.62±2.50	93.345	11.62±4.62	83.273
рН	7.22±0.20	7.21±0. 20		7.22±0. 20		7.22±0. 20	
alkalinity	207.25±8.97	77.25±2.60	62.726	77.56±2.77	62.576	77.31±2.71	62.697
DO	3.45±1.03	6.76±2.25		7.33±1.82		6.37±1.84	
samples °C	17.65 ±1.02	16.48±1.68		16.70±1.44		17.12±1.37	

Note: *concentration shown are mean ± standard deviation.

Evaluated the performance of pilot-scale downflow BAFs using 0.78 ± 0.60 mm activated carbon-based filtration media bed for nitrogen conversion and removal. The concentrations of the mean influent municipal wastewater approximately 3.16 ± 2.46 mg/L, 2.46 ± 2.0725 mg/L, 1.99 ± 1.76 mg/L, 10.46 ± 6.74 mg/L, and 32.12 ± 26.93 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 90.11%, 90.10%, 91.84%, 86.37%, 76.26%, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Where the mean effluent concentration was 0.31 ± 0.37 mg/L, 0.24 ± 0.31 mg/L, 0.16 ± 0.24 mg/L, 1.42 ± 0.95 mg/L, and 1.62 ± 4.49 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Furthermore, the pilot-scale downflow BAFs using activated carbon-based filtration media bed was

^{*}data shown are 33-day running mean.

able to nitrify 0.24 ± 0.21 kg NH₄+-N/m³.d, 0.19 ± 0.18 kg NO₂-N/m³.d, 0.15 ± 0.15 kg NO₃-N/m³.d, 0.78 ± 0.54 kg TKN/m³.d. Meanwhile, the pilot-scale downflow BAFs using activated carbon-based filtration media bed was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS, the mean influent concentrations approximately 63.25 ± 28.62 mg/L, 41.50 ± 12.86 mg/L, and 59.50 ± 48.59 mg/L, respectively for tCOD, sCOD, SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 91.89%, 91.47%, 94.24%, respectively for tCOD, sCOD, SS. Where the mean effluent concentration was 5.12 ± 0.99 mg/L, 3.53 ± 0.79 mg/L, 4.00 ± 1.92 mg/L, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.07 ± 2.42 kg tCOD/m³.d, 3.31 ± 1.06 kg sCOD/m³.d, and 5.72 ± 4.08 kg SS/m³.d, as showing in Table 5.2.

There have been evaluated the performance of pilot-scale downflow BAFs using 0.95±0.58 mm sandbased filtration media bed for nitrogen conversion and removal. The average influent concentrations approximately 3.16 ± 2.46 mg/L, 2.46 ± 2.0725 mg/L, 1.99 ± 1.76 mg/L, 10.46 ± 6.74 mg/L, and 32.12±26.93 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 87.74%, 89.84%, 90.28%, 84.82%, and 75.29%, respectively for NH_4^+-N , NO_2^--N , NO_3^--N , TKN. Where the mean effluent concentration was 0.38±0.44 mg/L, 0.25±0.32 mg/L, 0.19±0.27 mg/L, 1.58±0.97 mg/L, and 7.93±4.79 mg/L, respectively for NH₄+-N, NO₂-N, NO₃-N, TKN. Furthermore, the pilot-scale downflow BAFs using sand-based filtration media bed was able to nitrify 0.24±0.22 kg $NH_4^+-N/m^3.d$, 0.19 ± 0.18 kg $NO_2^--N/m^3.d$, 0.15 ± 0.15 kg $NO_3^--N/m^3.d$, 0.77 ± 0.54 kg $TKN/m^3.d$. Meanwhile, the pilot-scale downflow BAFs using sand-based filtration media bed was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS, the mean influent concentrations approximately 63.25±28.62 mg/L, 41.50±12.86 mg/L, and 59.50±48.59 mg/L, respectively for tCOD, sCOD, SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 91.60%, 90.96%, 93.34%, respectively for tCOD, sCOD, SS. Where the mean effluent concentration was 5.31±1.03 mg/L, 3.75±0.84 mg/L, 4.62±2.50 mg/L, respectively for tCOD, sCOD, SS. And the mean mass removal was $5.06\pm2.42 \text{ kg tCOD/m}^3.d$, $3.28\pm1.08 \text{ kg sCOD/m}^3.d$, and $5.66\pm4.03 \text{ kg SS/m}^3.d$, as showing in Table 5.2.

Evaluated the performance of pilot-scale downflow BAFs using 3.28±2.14 mm ceramic particle-based filtration media bed for nitrogen conversion and removal. The mean influent concentrations approximately 3.16±2.46 mg/L, 2.46±2.0725 mg/L, 1.99±1.76 mg/L, 10.46±6.74 mg/L, and 32.12±26.93 mg/L, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 85.17%, 88.32%, 89.03%, 83.69%, and 74.82%, respectively for NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, TKN. Where the mean effluent concentration was 0.46±0.46 mg/L, 0.28±0.34 mg/L, 0.21±0.29 mg/L, 1.70±1.00 mg/L, and 8.08±4.82 mg/L, respectively for NH₄+-N, NO₂--N, NO₃--N, TKN. Furthermore, the pilot-scale downflow BAFs using ceramic particle-based filtration media bed was able to nitrify 0.23 ± 0.22 kg NH₄+-N/m³.d, 0.18 ± 0.18 kg NO₂-N/m³.d, 0.15 ± 0.15 kg NO₃-N/m³.d, 0.76 ± 0.54 kg TKN/m³.d. Meanwhile, the pilot-scale downflow BAFs using ceramic particle-based filtration media bed was able to achieve a good reduction/removal of the carbonaceous, in terms of tCOD, sCOD, and SS, the mean influent concentrations approximately 63.25±28.62 mg/L, 41.50±12.86 mg/L, and 59.50±48.59 mg/L, respectively for tCOD, sCOD, SS. Where the experiments showed that with recirculation of the influent 100%, at HRT 12-hours, the mean removal efficiency has reached 90.51%, 90.51%, 83.27%, respectively for tCOD, sCOD, SS. Where the mean effluent concentration was 5.56 ± 0.97 mg/L, 3.93 ± 0.97 mg/L, 11.62 ± 4.62 mg/L, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.03 ± 2.43 kg tCOD/m³.d, 3.28 ± 1.05 kg sCOD/m³.d, and 5.05 ± 3.85 kg SS/m³.d, as showing in Table 5.2.

Table 5.2. Characteristics of the mean mass removal versus the removal efficiency by the nitrification and denitrification process during a period of testing conditions.

Parameter	BAFs us	le downflow ed activated based bed	BAFs use	le downflow d sand-based erial bed	Pilot-scale downflow BAFs used ceramic particle-based bed	
	Mass removal Kg/m³.d	Removal efficiency %	Mass removal Kg/m³.d	Removal efficiency %	Mass removal Kg/m³.d	Removal efficiency %
NH ₄ +-N	0.24±0.21	90.118	0.24±0.22	87.747	0.23±0.22	85.179
NO ₂ N	0.19±0.18	90.103	0.19±0.18	89.847	0.18±0.18	88.324
NO ₃ -N	0.15±0.15	91.849	0.15±0.15	90.284	0.15±0.15	89.030
TKN	0.78±0.54	86.379	0.77±0.54	84.826	0.76±0.54	83.692
tCOD	5.07±2.42	91.898	5.06±2.42	91.602	5.03±2.43	91.207
sCOD	3.31±1.06	91.478	3.28±1.08	90.965	3.28±1.05	90.514
SS	5.72±4.08	94.244	5.66±4.03	93.345	5.05±3.85	83.273

Note: *Mass removal shown are mean ± standard deviation.

The stoichiometry sCOD/TKN ratio describes the amount of organic carbon required to remove nitrate from the wastewater. It is essential to reflect the impact of the sCOD/TKN ratio on nitrogen elimination from wastewater. However, pretreatment of raw wastewater for SS elimination can result in the loss of OM, lowering the (sCOD/TKN ratio) of the wastewater. Adequate COD should be supplemented to fully denitrify the nitrate produced during the nitrogen reduction nitrification process. These experiments with three different sCOD/TKN ratios (2.8, 4.0, 4.5). Run 30 C/N ratio = 2.8 where a feed carbon and ammonium-nitrogen loading was 0.0483 Kg sCOD/m³.d, 0.0895 kg tCOD/m³.d, and 0.0058 kg NH₄+-N/m³.d. Run 25 C/N ratio = 4.0 where the loading was 0.0262 Kg sCOD/m³.d, 0.0344 kg tCOD/m³.d, and 0.0017 kg NH₄+-N/m³.d. Run 29 C/N ratio = 4.5 where the loading was 0.0202 Kg sCOD/m³.d, 0.0337 kg tCOD/m³.d, and 0.0011 kg NH₄+-N/m³.d.

The results of the experiments showed that all three filtration media used in the pilot-scale downflow BAFs with three different sCOD/TKN ratios (2.8, 4.0, 4.5) indicate that as the sCOD/TKN ratio increased, the nitrogen (NH_4^+ -N, NO_2^- -N, and NO_3^- -N) removal (%) decreased linearly, as well as, increased the sCOD/TKN ratio, the mass of the nitrogen removed (NH_4^+ -N, NO_2^- -N, and NO_3^- -N) decreased linearly. Correspondingly, the effluent of the nitrogen (NH_4^+ -N, NO_2^- -N, and NO_3^- -N) concentrations increased as the sCOD/TKN ratio increased.

The experimental results with three different sCOD/TKN ratios (2.8, 4.0, 4.5) indicate that as the sCOD/TKN ratio increased, the COD (sCOD, and tCOD) loading decreased, and COD (sCOD, and tCOD) removal (%) decreased, Correspondingly, the sCOD/TKN ratio decreased, the mass of the COD removed (sCOD, and tCOD) per unit volume (kg/m³.d), and removal (%) increased.

^{*}data shown are 33-day running mean.

5.4 General Conclusions of the Research

The main goal of the thesis was nitrogen conversion and removal by biological aerated filtration, as well as numerous working objectives derived from this main goal. Where the laboratory experiments by three identical pilot-scale downflow of BAFs using three different filtration media types, 0.78±0.60 mm activated carbon-based filtration media bed, 0.95±0.58 mm sand-based filtration media bed, and 3.28±2.14 mm ceramic particle-based filtration media bed as biomass support as attached growth zones in the treatment of municipal wastewater. The findings from the experiments on three pilot-scale downflow of BAFs during the monitoring period as follows:

- At ammonium-nitrogen loading rates approximately 0.0022±0.00 kg NH₄⁺-N/m³.d, with recirculation of the influent 100% at HRT 12-hours. The pilot-scale downflow BAFs using 0.78±0.60 mm activated carbon-based filtration media bed was able to nitrify 0.24±0.21 kg NH₄⁺-N/m³.d, with mean removal efficiency ammonium-nitrogen NH₄⁺-N at90.11%. The pilot-scale downflow BAFs using 0.95±0.58 mm sand-based filtration was able to nitrify 0.24±0.22 kg NH₄⁺-N/m³.d, with mean removal efficiency ammonium-nitrogen NH₄⁺-N at87.74%. The pilot-scale downflow BAFs using 3.28±2.14 mm ceramic particle-based filtration media bed was able to nitrify 0.23±0.22 kg NH₄⁺-N/m³.d, with mean removal efficiency ammonium-nitrogen NH₄⁺-N at 85.17%.
- According to the experimental results indicate that with recirculation of the influent 100% at HRT 12-hours, the activated carbon-based filtration media bed and sand-based filtration media bed have better capability than the ceramic particle-based filtration media bed in the nitrification and denitrification process. However, all three filtration media used in the pilotscale downflow BAFs were able to reach the mean final effluent concentration of ammoniumnitrogen NH₄+-N less than 0.47 mg/L.
- At organic loading rates approximately 0.0434±0.01kg tCOD/m³.d, 0.0285±0.00 kg sCOD/m³.d, and 0.0931±0.17 kg SS/m³.d, with recirculation of the influent 100% at HRT 12-hours. The pilot-scale downflow BAFs using 0.78±0.60 mm activated carbon-based filtration media bed was able to achieve good reduction/removal of carbonaceous at 91.89%, 91.47%, 94.24%, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.07±2.42 kg tCOD/m³.d, 3.31±1.06 kg sCOD/m³.d, and 5.72±4.08 kg SS/m³.d. The pilot-scale downflow BAFs using 0.95±0.58 mm sand-based filtration media bed was able to achieve good reduction/removal of carbonaceous at 91.60%, 90.96%, 93.34%, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.06±2.42 kg tCOD/m³.d, 3.28±1.08 kg sCOD/m³.d, and 5.66±4.03 kg SS/m³.d. The pilot-scale downflow BAFs using 3.28±2.14 mm ceramic particle-based filtration was able to achieve good reduction/removal of carbonaceous at 90.51%, 90.51%, 83.27%, respectively for tCOD, sCOD, SS. And the mean mass removal was 5.03±2.43 kg tCOD/m³.d, 3.28±1.05 kg sCOD/m³.d, and 5.05±3.85 kg SS/m³.d.

Thus, in conclusion, that all the three-filtration media-based bed which proposed in this research their efficiency of elimination of the nitrogen by the nitrification and denitrification process extremely well. However, of the three-filtration, media-based bed types, the pilot-scale downflow BAFs which used a 0.78±0.60 mm activated carbon-based filtration media bed showed the highest percentage of nitrogen removal efficiency and mass nitrogen removal.

5.5 The Main Achievements of the Research

The main achievements of the research are:

- It was revealed that the reactors which used proposed filtration media-based beds very
 effective in nitrogen conversion and removal, where could be utilized as attached growth
 zones in the treatment of municipal wastewater. Where it is easily obtained, not
 manufactured and proprietary, locally available, low cost.
- The larger sized material 3.28±2.14 mm ceramic particle-based filtration media bed, showed inferior in SS removal results, compared with 0.78±0.60 mm activated carbon-based filtration media bed and a 0.95±0.58 mm sand-based filtration media bed.
- All the proposed filtration media-based beds were able to effectively nitrify, with a mean removal efficiency of the ammonium-nitrogen NH₄⁺-N was more than 90%.
- All the proposed filtration media-based beds were effective in reduction/removal of the carbonaceous with a mean removal efficiency of COD more than 90%, and SS between 83% to 94%.
- With three different sCOD/TKN ratios (2.8, 4.0, 4.5) specify that as the sCOD/TKN ratio increased, the nitrogen removal (%) decreased linearly, as well as, the sCOD/TKN ratio increased, the mass of the nitrogen eliminated per unit volume (kg/m³.d) decreased linearly. Correspondingly, the effluent of the nitrogen level (mg/L), increased as the sCOD/TKN ratio increased.

5.6 Recommended Future Research

The following subjects are considered a normal continuation of the research described in the present thesis:

- Study the temperature effect on the performance of the nitrogen removal in the BAFs;
- Study effect filtration media type on effective phosphorus removal in the BAFs;
- Study the nitrification and phosphorus precipitation in the BAFs;
- Investigate the effect of the COD:N:P ratio on the C/N/P ratios needed for successful nutrient elimination in the BAFs;
- Study the optimum oxygen level, HRT, and loading rates for instantaneous nitrification, denitrification, and P elimination in the BAFs.

5.7 Publications

Following the presented research works there were published, presente or submitted several articles as follows:

- Already published:
- 1. Faskol, A. S., Racovițeanu, G. (2020). Nitrification and denitrification process employing three different sunken materials types in biological aerated filter BAFs. *International Journal of Engineering Research And Management (IJERM)*. (07(10)), pp. 1-5.
- 2. Faskol, A. S., Racovițeanu, G. (2021). Technology review of the biological aerated filter systems BAFs for removal of the nitrogen in wastewater. *IN: Proceedings of the 7th Conference of the Sustainable Solutions for Energy and Environment (Bucharest, Romania)*. IOP Conf. Series: Earth and Environmental Science 664 (2021) 012106.

- 3. Faskol, A. S., Racovițeanu, G. (2021). Effect of DO, Alkalinity and pH on Nitrification Using Three Different Sunken Materials Types in Biological Aerated Filter BAFs. *IN: Proceedings of the 7th Conference of the Sustainable Solutions for Energy and Environment (Bucharest, Romania)*. IOP Conf. Series: Earth and Environmental Science 664 (2021) 012079.
 - Under Publication:
- 4. Faskol, A. S., Racovițeanu, G., Vulpasu, E., (2021). The Ratio Carbon/Nitrogen Influence on the Nitrification Process in Biological Aerated Filter.
- 5. Faskol, A. S., Racovițeanu, G., Vulpasu, E., (2021). Impact of three different filtration media types on the elimination of ammonium-nitrogen in BAFs.
- 6. Faskol, A. S., Racovițeanu, G., Vulpasu, E., (2021). Combined COD and ammonium-nitrogen elimination a BAFs comparison between three different filtration media types.

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