

Removal of ammonium ions from wastewater A short review in development of efficient methods

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ABSTRACT: Ammonium ions wastewater pollution has become one of the most serious environmental problems today. The treatment of ammonium ions is a special concern due to their recalcitrance and persistence in the environment. In recent years, various methods for ammonium ion removal from wastewater have been extensively studied. This paper reviews the current methods that have been used to treat ammonium ion wastewater and evaluates these techniques. These technologies include ion exchange, adsorption, biosorption, wet air oxidation, biofiltration, diffused aeration, nitrification and denitrification methods. About 75 published studies (1979-2015) are reviewed in this paper. It is evident from the literature survey articles that ion exchange, adsorption and biological technology are the most frequently studied for the treatment of ammonium ion wastewater.

Keywords: Ammonium wastewater, Removal methods, Review, Toxicity, Treatment

INTRODUCTION

Ammonium ions are the primary form of widespread nitrogen pollution in the hydrosphere and caused a remarkable increase of oxygen demand and biological eutrophication in local by aquatic sources (Sprynsky *et al.*, 2005) and results proved that increased in concentration of this beyond a permissible cause damage to aquatic life (For example, ammonia is toxic to fish and other forms of aquatic life in very low concentration, about 0.2 mg/L) (Randall and Tsui, 2002; Haralambous *et al.*, 1992). Thus the major concern regarding ammonia toxicity must be in aquatic systems, particularly in regions of high human habitation and/or large numbers of farm animals (Ip *et al.*, 2001).

Hence, the removal of ammonium from municipal and industrial wastewater prior to discharge is now

obligatory. Many methods are being used to remove ammonium ions. The present review article deals with the current techniques for the ammonium ions removal from wastewater (Fig. 1). Their advantages and limitations in application are also evaluated.

Toxicity and source of ammonium ions

Molecular nitrogen (N_2) present in the earth atmosphere has to be reduced to ammonia (NH_3) by nitrogen-fixing bacteria living independently in the soil or in the root of leguminous plants before it is utilized by humans. Ammonia dissolves in water to form ammonium ions (NH_4^+) and this form of reduced nitrogen is assimilated into amino acids and other nitrogen-containing molecules. In aqueous solutions, ammonia is a base (any compound accepting hydrogen ions) forming a conjugated pair with the ammonium ion, according to the reversible reaction:

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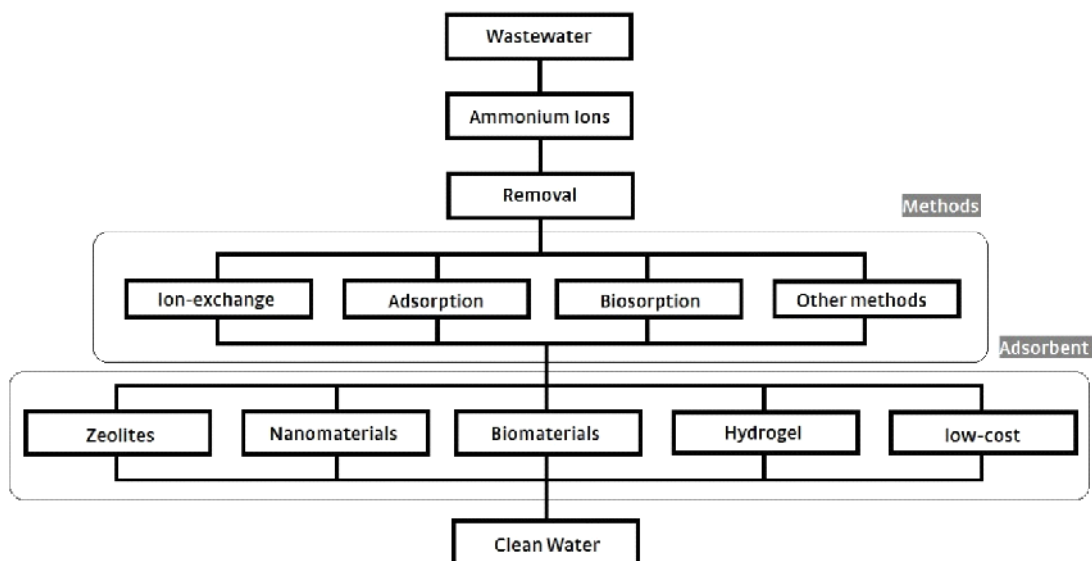


Fig. 1: Flowchart of ammonium ions removal from wastewater



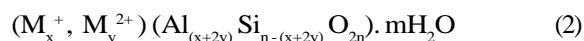
The pK_a of the reaction is 9.3, which indicated that at this pH value, the concentration of the ionized (NH_4^+) and unionized (NH_3) forms is equal. When the pH of the solution is less than 9.3, hydrogen ions are incorporated to ammonia to yield ammonium ions. Therefore, at physiological plasma and intracellular pH values, virtually only the protonated moiety (NH_4^+) is present in aqueous solutions (Graham and MacLean, 1992; Katz *et al.*, 1986; Adeva *et al.*, 2012).

Ammonia has a hazardous and toxic effect on human health and biotic resources also, only if the intake becomes higher than the capacity to detoxify and predefined permissible limits. If ammonia is administered in the form of its ammonium salts, the effects of the anion must also be taken into account. With ammonium chloride, the acidotic effects of the chloride ion seem to be of greater importance than those of the ammonium ion (Ryer Powder, 1991). At a dose of more than 100 mg/kg of body weight per day (33.7 mg of ammonium ion per kg of body weight per day), ammonium chloride influences metabolism by shifting the acid–base equilibrium, disturbing the glucose tolerance, and reducing the tissue sensitivity to insulin (Lamm *et al.*, 1999; Sadegh *et al.*, 2014a). So, Ammonium ion and various ammonium product based azo dyes compounds are toxic and hazardous to both the environment and human body, hence to

remove these toxic pollutants and dyes several adsorbents are developed and these adsorbents has an excellent removal capacity (Gupta *et al.*, 2015).

Removal of ammonium ions methods

Ion-exchange process has been used in various fields in recent years (see Table 1), including the ammonium ions removal from wastewater due to their many unique properties, such as high treatment capacity, high removal efficiency, low cost and fast kinetics (Cooney *et al.*, 1999; Kang *et al.*, 2004). One of the most used compound as ion-exchanger in removal ammonium was zeolites (urkovi *et al.*, 1997). Zeolites are highly porous alumino-silicates with different cavity structures that consist of a three dimensional framework and a negatively charged lattice (Bekkum *et al.*, 1991). The negative charge is balanced by cations which are exchangeable with certain cations in solutions. High ion-exchange capacity and relatively high specific surface areas, and more importantly their relatively low prices, make zeolites attractive adsorbents in the removal of ammonium ions (Sprynsky *et al.*, 2005; Haralambous *et al.*, 1992; Blocki, 1993; Ning *et al.*, 2008; Zhao *et al.*, 2007). The general formula of a zeolite is:



Where M^+ and M^{2+} are monovalent and divalent cations such as Na^+ , K^+ , and Ca^{2+} , Mg^{2+} , Ba^{2+} ,

Table1: The summary of reports on ammonium ions removal by ion exchange method

Adsorbent	Maximum adsorption capacity (mg/g)	surface area (m ² /g)	Removal percentage (%)	Conditions	Report by
Natural Turkish zeolite	NA	NA	75-83	pH: 4-9, C ₀ : 60 mg/L adsorbent dosage: 0.4 g shaking time: 60 min. temperature: 21 °C	Saltali <i>et al.</i> , (2007)
Natural zeolite	0.38	2.0+1.0	NA	pH: 7-7.5, C ₀ : 80 mg/L adsorbent dosage: 10 g shaking time: 20 min. temperature: 20 °C	Demir <i>et al.</i> , (2002)
Natural zeolite	1.64	NA	NA	pH: NA; C ₀ : 100 mg/L shaking time: 45 min. adsorbent dosage: 0.5 g temperature: 25 °C	Sprynskyy <i>et al.</i> , (2005)
Natural Iranian zeolite	NA	NA	91.5	pH: 7, C ₀ : 0.3 mg/L adsorbent dosage: 1 g shaking time: 90 min. temperature: 25±1 °C	Malekian <i>et al.</i> , (2011)
Natural Australian zeolite	1.5	NA	75-95.6	pH: to 11, C ₀ :8.79 mg/L adsorbent dosage: 1g shaking time: 65 min. temperature: 20 °C	Cooney <i>et al.</i> , (1999)
Clinoptilolie zeolite	1.05	NA	95-98	pH: 7, C ₀ :101 mg/L adsorbent dosage: 30 g shaking time: 25 min. temperature: 20 °C	Rahmani <i>et al.</i> , (2004)
Zeolites namely strong	74.7	NA	88-99.9	pH: 7.35-7.77, C ₀ : 580 mg/L adsorbent dosage: 0.7-27.3 g temperature: 20 °C shaking time: 80 min	Malovanyy <i>et al.</i> , (2013)
Yemeni natural zeolite	11	35.85	99	pH: 8, C ₀ : 80 mg/L adsorbent dosage: 1.2 g shaking time: 30 min. temperature: 80 °C	Alshameri <i>et al.</i> , (2014)
Zeolite mesolite	49	NA	70	pH: 6-7, C ₀ : 400 mg/L adsorbent dosage:1-5 g shaking time: 25 min. temperature: °C	Thornton <i>et al.</i> , (2007a)
Zeolite mesolite	55	NA	95	pH: 6-8, C ₀ : 50 mg/L adsorbent dosage: 1 g shaking time: 120 min. temperature: 25 °C	Thornton <i>et al.</i> , (2007b)

NA: not available, C₀: initial concentration (mg/L)

respectively. They are called exchangeable cations. Al³⁺ and Si⁴⁺ are known as structural cations, and they

make up the framework of the structure with O (Saltalı *et al.*, 2007).

Natural zeolite is the most abundant and frequently zeolite, which shows a very high specific selectivity for certain pollutants (Erdem *et al.*, 2004; Meshko *et al.*, 2001; Peri *et al.*, 2004; Sarioglu, 2005; Kesraoui-Ouki *et al.*, 1994). Demir *et al.*, (2002) studied the ammonium removal from aqueous solution by ionexchange using packed bed natural zeolite and reported that the resin column was exhausted by down flow at 10, 25, 50 and 75 bed volume (BV)/h, until effluent ammonium concentration of more than 10 mg/L NH_4^+ was achieved. The results indicate that conditioning of the zeolite improves the ion-exchange capacity and the smaller particle size also causes a higher ion-exchange capacity due to greater available surface area. The actual ion-exchange capacity of the conditioned fine (-1.00+0.125) and coarse (-2.00+1.00) mm clinoptilolite was found to be 0.57 and 0.38 meq/g zeolite, respectively. Sprynskyy *et al.*, (2005) studied ammonium ions removal from synthetic aqueous solutions by raw and pretreated natural zeolite. Sprynskyy *et al.* assessed the ability of zeolitic tuffs such as mordenite to remove 1000 ppm aqueous ammonia. Malekian *et al.*, (2001) used the natural Iranian zeolite is a suitable ion-exchange for NH_4^+ ion removal and consequently has potential as a controlled-release NH_4^+ fertilizer. Cooney *et al.*, (1999) revealed that the highest percentage ammonium removal efficiency and high removal rate using natural Australian zeolite. Rahmani *et al.*, (2004) studied the efficiency of an ammonium ions removal by Clinoptilolite zeolite and reported Clinoptilolite may be effectively applied in ammonium ions. Malovanyy *et al.*, (2013) studied the application of four types of ion-exchange materials in packed bed columns, namely strong and weak acid cation exchange resins and natural and synthetic zeolites. Malovanyy *et al.*, (2013) reported that ammonium ions removal efficiency in most cases was higher than 95% which allows using ion-exchange processes. Recently Alshameri *et al.*, (2014) reported the potential use of Yemeni (Al-Ahyuq) natural zeolite on the effectiveness of ammonium ion (NH_4^+) removal and this research indicates that the modified Yemeni zeolite has significant potential as an economic and effective adsorbent material for ammonium removal from aqueous solutions.

Thornton *et al.*, (2007a, b) reported that using Mesolite ion exchange media for ammonium (NH_4^+) removal from solution gives an excellent result and high removal rate also. Zhao *et al.*, (2007) reported that magnetic zeolite could be used for the removal of

ammonium due to the good adsorption performance and easy separation method from aqueous solution.

According to Table 2, adsorption is now recognized as an effective and economic method for ammonium ions removal (Moradi, 2011). The adsorption process offers flexibility in design and operation; in fact it will produce high-quality treated effluent in many cases. In addition, since adsorption is sometimes reversible, adsorbents can be regenerated by suitable desorption process (Rosenfeld, 1979). Recently Moradi and Zare, (2013) reported an attempt to examine the feasibility of removing ammonium ions from aqueous solutions by using multiwalled carbon nanotube (MWCNT) and reported removal percentage and adsorption of ammonium ions on to CNTs was highly efficient. Wang *et al.*, (2014) reported palygorskite (PGS) nanocomposite as adsorbent were highly efficient, the results suggests that the novel composite hydrogel (PGS) was suitable to the adsorption of NH_4^+ from aqueous solution, especially in the agricultural sector where nitrogen-laden carrier material can be reused as a multifunctional slow-release fertilizer to enhance soil fertility and help the amelioration of soils. Searching for low-cost and easily available adsorbents to remove ammonium ions has become a main research focus, Hence till date, many of studies on the use of low-cost adsorbents have been published, for example, Maranon *et al.*, (2006) investigated the Romanian volcanic tuff is able to uptake ammonium ions from an aqueous solution, showing high specific selectivity for this cation and conclude that Romanian volcanic tuff may be used successfully as an alternative adsorbent in the treatment of wastewater containing ammonium at concentrations of up to 100 mg/L. Ma *et al.*, (2011) studied the removal of ammonium ion from water by fertilizer controlled release agent prepared from wheat straw.

In recent years, another adsorbent used in adsorption processes is natural adsorbent and the most efforts of researchers have been made to find the adsorbents with higher efficiency and lower cost (Sartape *et al.*, 2010; González *et al.*, 2011; Sadegh *et al.*, 2014b). Natural adsorbent such as natural clays are one of the considerable and applicable sorbents due to their low-cost, availabilities and easy applications in the removal of ammonia contaminants from environment (Wang *et al.*, 2014). Another Natural adsorbent used in adsorption was zeolite (Balci and Dinçel, 2002; Otal *et al.*, 2013; Huo *et al.*, 2012; Huang *et al.*, 2010; Vassileva and Voikova, 2009; Arslan and

Table 2: The summary of reports on ammonium ions removal by adsorption method

Adsorbent	Maximum adsorption capacity (mg/g)	surface area (m ² /g)	Removal percentage (%)	Conditions	Report by
Carbon nanotubes	17.05	NA	97	pH: 7-11, C ₀ : 100 mg/L adsorbent dosage: 0.05g shaking time: 35 min. temperature: 25 °C	Moradi and Zare, (2013)
Nano-palygorskite/nano composite	237.6	NA	60	pH: 4-8, C ₀ : 100 mg/L adsorbent dosage: 0.2 g shaking time: 15 min. temperature: 20-30 °C	Wang <i>et al.</i> , (2014)
Low cost - Romanian volcanic tuff	19	52.02±0.28	83	pH: 7, C ₀ : 100 mg/L adsorbent dosage: 1 g shaking time: 180 min. temperature: 22 °C	Maranon <i>et al.</i> , (2006)
Low cost -wheat straw	148.7	NA	75	pH: 4-8, C ₀ : 100 mg/L adsorbent dosage: 1 g shaking time: 4h. temperature: 22 °C	Ma <i>et al.</i> , (2011)
Zeolite	13.73	NA	70	pH: 8.2, C ₀ : 18.5 mg/L adsorbent dosage: 3g shaking time: 25 min. temperature: 20 °C	Otalet <i>et al.</i> , (2013)
Zeolite clinoptilolite	NA	NA	98.8	pH: 8.2, C ₀ : 18.5 mg/L adsorbent dosage: NA shaking time: 3h. temperature: 30 °C	Huo <i>et al.</i> , (2012)
Zeolite natural Chinese	9.41	NA	95	pH: 8, C ₀ : 80 mg/L adsorbent dosage: 24g shaking time: 180 min. temperature: 25 °C	Huang <i>et al.</i> , (2010)
Zeolite clinopilot	12.29	26.0	85	pH: 6, C ₀ : 175 mg/L adsorbent dosage 0.25g shaking time : 60 min. temperature: 20 °C	Vassileva and Voikova, (2009)
Zeolite 13X	4.80	NA	90	pH: 7, C ₀ : 25 mg/L adsorbent dosage: 0.5g shaking time: 200 min. temperature: 23 °C	Arslan and Veli, (2012)
Zeolite	12	NA	NA	pH: 7, C ₀ : 150 mg/L adsorbent dosage: 50g shaking time: 2000 min. temperature: 25 °C	Cincotti <i>et al.</i> , (2001)
Zeolite	9.479	NA	89	pH: 5.23, C ₀ : 30 mg/L adsorbent dosage: 1g shaking time: 480 min. temperature: 95-100 °C	Zheng <i>et al.</i> , (2008)

Natural zeolite	3.11	NA	90	pH: 7, C_0 : 30 mg/L adsorbent dosage: 0.05g shaking time: 30 min. temperature: 25 °C	Alshameri <i>et al.</i> , (2014)
Polymer-hydrogel PVA	42.74	NA	70	pH: 3-8, C_0 : 100 mg/L adsorbent dosage: 1 & 1.8g shaking time: 35 min. temperature: 30 °C	Zheng <i>et al.</i> , (2011)
Aerobic granular sludge activated	40	NA	65	pH: 7, C_0 : 30 mg N/L adsorbent dosage: 0.2mg shaking time: 35 min. temperature: 20-30 °C	Bassin <i>et al.</i> , (2011)
Aerobic granules	24.5	NA	NA	pH: 7, C_0 : 300 mg/L adsorbent dosage: 1mg shaking time: 60min. temperature: 30 °C	Yu <i>et al.</i> , (2014)
Biosorbent	NA	NA	97.5	pH: 5, C_0 : 300 mg/L adsorbent dosage: 5mg shaking time: 360min. temperature: 25 °C	Suneetha and Ravindhranath, (2012)
Municipal sludge	NA	NA	89	pH: 10, C_0 : 300 mg/L adsorbent dosage: 5 mg shaking time: 2min. temperature: 25 °C	Ismail <i>et al.</i> , (2010)

NA: not available, C_0 : initial concentration (mg/L)

Veli, 2012; Cincotti *et al.*, 2001; Zheng *et al.*, 2008; Alshameri *et al.*, 2014).

There are several literature reported on the adsorption of ammonium ion by other adsorbent. Zheng *et al.*, (2011) reported the application of hydrogel polyvinyl alcohol (PVA) surface in the removal of ammonium ion. Bassin *et al.*, (2011) used aerobic granular sludge, activated sludge and anammox granules as adsorbent in ammonium adsorption. Yu *et al.*, (2014) also use aerobic granules, Suneetha and Ravindhranath, (2012) were used adsorbents derived from powders of leaves, stems or barks of some plants. Also Ismail *et al.*, (2010) reported adsorption ammonium on municipal sludge.

Biosorption of ammonium ions from aqueous solutions is a relatively new process that has been confirmed a very promising process in the removal of ammonium ion contaminants. The major advantages of biosorption are its high effectiveness in reducing the ions and the use of inexpensive biosorbents. Biosorption processes are particularly suitable to treat dilute ammonium wastewater. Typical biosorbents can be derived from three sources as follows (Apiratikul and Pavasant, 2008): (1) non-living biomass such as

bark, lignin, shrimp, krill, squid, crab shell, etc.; (2) algal biomass; (3) microbial biomass, e.g. bacteria, fungi and yeast. Different forms of inexpensive, such as *Posidonia oceanica* (*P. oceanica* fibers) (Jellali *et al.*, 2011), activated sludge (Ren *et al.*, 2011), heterotrophic bacteria (Zhang *et al.*, 2013). Jellali *et al.*, (2011) reported that the low-cost, availability and high adsorption capability of the *P. oceanica* fibers make them a promising and potentially attractive biosorbents for removal of ammonium ion.

There is a large number of research works on the ammonium biosorption using biological technology (Bouwer and Crowe, 1988). Examples of recent reports include the wet air oxidation (WAO) (Bernardi *et al.*, 2012), ozonation (Schroeder *et al.*, 2011; Tanaka and Matsumura, 2003), intermittent flow biofilter (IBF) (Sabbah *et al.*, 2013), biofiltration (Azman *et al.*, 2014), nitrification and denitrification (Feng *et al.*, 2012; Qiao *et al.*, 2010; Huang *et al.*, 2013) (Table 3). The summary of reports on ammonium ions removal by Biosorption method is indicated in Table 3.

Other methods of ammonia removal include: ammonia volatilization (Valero and Mara, 2007), sharonprocess (Hellinga *et al.*, 1998), simultaneous

Table 3: The summary of reports on ammonium ions removal by Biosorption method

Adsorbent	Maximum adsorption capacity (mg/g)	surface area (m ² /g)	Removal percentage (%)	Conditions	Report by
Posidoniaoceanica (L.) fibers	1.8	NA	NA	pH: 6, C ₀ : 50 mg/L adsorbent dosage: 3g shaking time: 40min. temperature: 18± 02 °C	Apiratikul and Pavasant, (2008)
Activated sludge	88.4	31.84	95	pH: 6, C ₀ : 20 mg/L adsorbent dosage: 3 g shaking time: 40 min. temperature: 25 °C	Ren <i>et al.</i> , (2011)
Microbacterium sp. strain SFA13	NA	NA	91.8	pH: 7, C ₀ : 80 mg/L adsorbent dosage: 10 g shaking time: 60 min. temperature: 5 °C	Zhang <i>et al.</i> , (2013)
Supported Pt catalysts	NA	NA	97.5	pH: 8, C ₀ : 1700, 1400, 60 mol /L adsorbent dosage: ... g shaking time: 6 h. temperature: 200 °C	Bernardi <i>et al.</i> , (2012)
Ozone	NA	NA	99	pH: 6.5-8.5, C ₀ : 1700, 1400, 0.06 mg/L adsorbent dosage: 1 g shaking time: 6 h. temperature: 27 °C	Schroeder <i>et al.</i> , (2011)
Ozone and BAC	8	NA	81.3	pH: 7 – 7.5, C ₀ : 35.5 & 181 mg/L adsorbent dosage:1 g shaking time: 3.5 Day. temperature: 25 °C	Baozhenet <i>al.</i> , (1989)
Ozone	0.0009– 0.445	85	NA	pH: 3, C ₀ : 1700, 1400, 701 mg/L adsorbent dosage: 1 g shaking time: 15 h. temperature: 20 °C	Tanaka and Matsumura, (2003)
Macro-algae	0.03	NA	70	pH: 5-9, C ₀ : 1.8-2.2 mg/L adsorbent dosage: 1 g shaking time: 60 and 120day. temperature: 26-27 °C	Sabbah <i>et al.</i> , (2013)
GAC-sand dualmedia filter	NA	NA	35-42	pH: 7.2, C ₀ : 1 mg/L adsorbent dosage: 3.06 mg shaking time: 20min. temperature: 10-30 °C	Feng <i>et al.</i> , (2012)
Novel acryl biofilm carrier material	NA	NA	98.7	pH: 6.8-7.8, C ₀ : 20 mg/L adsorbent dosage: 3.06 mg shaking time: 20 min. temperature: 5-30 °C	Qiao <i>et al.</i> , (2010)
Ammonia volatilization	NA	NA	90-99	pH: 10.1, C ₀ :6.13 mg/L adsorbent dosage: 0.5 mg shaking time: 36 h. temperature: 17.1 °C	Valero and Mara, (2007)

NA: not available, C₀: initial concentration (mg/L)

removal (Rahimpour and Mottaghi, 2009; Lv *et al.*, 2013; Tang *et al.*, 2014), and diffused aeration (Patoczka and Wilson, 1984).

Remarks of removal methods

Ion exchange has been widely applied for the removal of ammonium ions from wastewater. However, ion-exchange resins must be regenerated by chemical reagents when they are exhausted. This can cause serious secondary pollution; more over it is expensive, especially when it is treating a large amount of wastewater containing ammonium ions in low concentration, so they cannot be used at large scale. Adsorption is a recognized method for the removal of ammonium ions from low concentration wastewater containing ammonium ions. The high cost of CNTs limits its use in adsorption. A variety of low-cost adsorbents have been developed and tested to remove ammonium ions. However, the adsorption efficiency depends on the type of adsorbents. Biosorption of ammonium ion from aqueous solutions is a relatively new process that has proved very promising tool for the removal of ammonium ion from aquatic sources.

Although all above techniques can be employed for the treatment of ammonium ions wastewater, it is important to mention that the selection of the most suitable treatment techniques and adsorbent depends on the initial metal concentration, the component of the wastewater, capital investment and operational cost, plant flexibility and reliability and environmental impact, etc.

CONCLUSION

The presence of ammonium ions in wastewater is a major concern for environment conservation and human health. Until now, the process of removal of these ions has not reached the optimum conditions. To increase stringent environmental regulations more and more, a wide range of treatment technologies such as ion-exchange, adsorption, biosorption, wet air oxidation, biofiltration, diffused aeration, nitrification and denitrification methods have been developed for ammonium ions removal from wastewater. It is evident from the literature survey of 75 articles that ion-exchange, adsorption and biological technology are the most frequently studied for the treatment of ammonium ions wastewater. Ion-exchange processes have been widely used to remove ammonium from wastewater. Adsorption by low-cost adsorbents and biosorbents is recognized as an

effective and economic method for low concentration ammonium wastewater treatment.

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