Removal of ammonia from polluted waters using biosorbents derived from powders of leaves, stems or barks of some plants

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ABSTRACT

Powders of leaves and barks of Phyllanthus niruri, Annona squamosa, Calotropis gigantean, Tridax procumbens, Morinda tinctoria and Azadirachta indica have been found to have strong affinity towards ammonia. The sorption abilities of these bio-products in controlling the ammonia pollution in waste waters have been studied with respect various physicochemical parameters such as pH, sorption concentration and equilibration time. % of removal of ammonia is pH sensitive and also depends on sorption concentration and time of equilibration. More than 90% of ammonia can be removed using these bio-sorbents at pH: 5 and at optimum equilibration time and sorbent concentrations. The minimum sorbent dosage needed for the maximum removal of ammonia is found considerably less for barks powders than for powders of corresponding leaves. Anions like Chlorides, Fluorides, Sulphate, Phosphate and Carbonates are interfered marginally while cations like Ca$^{2+}$, Cu$^{2+}$, Zn$^{2+}$ and Mg$^{2+}$ interfered to some extent and however, the % of extractability of ammonia has never come down 70.0%. The adoptability of the methodologies developed in this work are tested with diverse samples collected from industrial effluents and naturally existing polluted lakes and it is found that the procedures are remarkably successful.

Key words: Ammonia, pollution control, bio-sorbents, applications.

INTRODUCTION

Ammonia is present in most waters as a result of the biological degradation of nitrogenous compounds [1-4]. Common sources of ammonia pollution include industrial wastes, agricultural input and sewage effluents. Ammonia nitrogen discharges from these sites, subsequently gains entrance into water resources and causes threat to aquatic life. Other potential sources of Ammonia pollution are the animal feeds, food additives, cleaning agents and effluents from the
industries manufacturing fibers, plastics, explosives, paper, and rubber; Further ammonia is also a metabolic by-product of fish [5].

Ammonia is a serious toxicant to aquatic organisms and is recognized in as an important factor to be considered when establishing water quality criteria for freshwaters [3,6]. Ammonia exists in water either in unionized ammonia (NH$_3$) or ionized ammonia (NH$_4^+$) depending up on the pH and temperature conditions. The toxicity of ammonia to aquatic organisms is due to the unionized fraction (NH$_3$) of ammonia. Ammonia causes headache, insomnia, nausea, diarrhea and a failure in glucose tolerance in animals and human beings [7-9]. There is evidence that ammonia targets cardiac tissue [10,11]. Ammonia causes metabolic toxicity and interferences with energy metabolism in the brain [12]. It is a strong cell poison and can cause damage to the gills of fish even at levels of 0.25ppm. Other deleterious effects of ammonia pollution include accelerated eutrophication of lakes, dissolved oxygen depletion in receiving waters and fish toxicity [7].

The maximum limit of ammonia set by the European Association for drinking water is approximately 0.5 ppm and also a guide level is given as 0.05ppm [13, 14]. The raw water with high ammonia concentration must therefore be treated before it reaches the consumer and also the wastewater discharge into the receiving water.

The most widely used methods for removing ammonia from wastewater are air stripping, ion exchange, and biological nitrification and de-nitrification.[15]. Biological nitrification and denitrification methods are limited to a minimum 5 ppm of concentration due to the formation of undesirable chemical compounds. The ion exchange methods [16-21] are preferred over the other methods since they are stable, suit automation and quality control and are easy to maintain. Clinoptilolite zeolites are used in the control of ammonia [21].

Recent interest is also focused on the using Microwave radiation for the control of ammonia [21-25]. New concepts of microbial treatment processes for the removal of ammonia have been discussed by Schmidt I et al (2003)[26]. M.S. Çelik et al (2001) studied the removal of ammonia by natural clay minerals fusing fixed and fluidized bed column reactors [27].

These methods involve large expenditure and so investigations are being made to explore the sorption potentialities of bio-wastes of flora and fauna origin as alternative methods in controlling pollution [28-32]. V. Parimala et. al (2007) investigated the removal of ammonia using low cost agricultural wastes [32]. Duck weeds [28], neem products [29] and low cost agricultural wastes [30, 32] have been explored for the removal of ammonia from polluted waters. These methods based on the use of waste materials as bio-sorbents are proving to be potential alternative to the existing costly methods for the removal of ammonia.

The endeavor of the present work is to explore the sorption potentialities of thermally activated powders of leaves and barks of some plants in controlling the concentrations ammonia in polluted waters by studying the sorption characteristics of these bio-sorbents under various physicochemical parameters such as pH, sorbent concentration and time of equilibration.
MATERIALS AND METHODS

(A) Chemicals: All chemicals used were of analytical grade.
1. 500 ppm stock solution of Ammonia was prepared by dissolving suitable amounts of Ammonium chloride in double distilled water and is suitably diluted as per the need.
2. Nessler’s reagent: 35gs of Potassium Iodide and 4% Mercuric Chloride were dissolved in 100 ml of double distilled water with constant stirring until a slight red precipitate remains. To this , solution of NaOH (120gms of NaOH + 250 ml of double distilled water ) was added and made up to 1lit with double distilled water. A little more Mercuric Chloride solution was added until there was a permanent turbidity. The mixture was allowed to stand for one day and decant from the sediment. The solution was kept in stoppered dark colored bottle.

(B) Adsorbents:
Thermally activated powders of leaves, stems or barks of many plants have been employed in this work. It is found that leaves, stems or barks of Phyllanthus niruri, Annona squamosa, Calotropis gigantean, Tridax procumbens, Morinda tinctoria and Azadirachta indica have been found to have affinity towards ammonia and hence the methodologies presented here, pertain to these sorbents only.

Phyllanthus Niruri                Annona squamosa                      Calotropis gigantean

Tridax procumbens               Morinda tinctoria                      Azadirachta indica

Phyllanthus Niruri is a herbal plant belonging to Phyllanthaceae family and is found in Central and Southern India; it is found to have many therapeutic values in curing jaundice, diabetes, dyspepsia, ulcers, sores, swellings, ophthalmia and chronic dysentery. Annona squamosa is a small well-branched shrub that bears edible fruits called sugar-apple; belongs to Annonaceae family and grows well in lower altitudes. Calotropis gigantean is a species of Calotropis, native
to South Asian countries and it belongs to Apocynaceae family. It is a large shrub growing up to 3-4 meters tall and it possesses wound healing ability. *Tridax procumbens* is a species of flowering plant in the daisy family and is best known as a widespread weed and pest plant. It grows in tropical, subtropical and mild temperate regions worldwide. It possesses medicinal uses especially for diabetic treatment. The oral administration of leaf extracts at doses of 200 mg kg⁻¹ lead to a significant blood glucose reduction. This laid the foundation to study the active compounds of such anti-diabetic plants that are responsible for the hypoglycemic activities. It also proves the traditional claim of Mandesh region with regard to *Tridax procumbens* for its anti-diabetic activity [33]. *Morinda tinctoria*, commonly known as Aal or Indian Mulberry is a species of flowering plant and belongs to Rubiaceae family and is native to South Asia. It is an evergreen shrub growing to 5-10 m tall. The plant is extensively cultivated in India and its leaves and roots are used in traditional system of medicine, as astringent, deobstrent, emmengogue and to relive pain in the gout [34]. It is reported to have anticonvulsant activity [35]. *Azadirachta indica*, or Neem Tree, is an evergreen tree native to Southeast Asia and it belongs to Meliaceae family.

**Sorbert Preparation:** The leaves or stems or barks of *Phyllanthus Niruri*, *Tridax procumbens*, *Morinda tinctoria*, *Azadirachta indica*, *Annona squamosa* and *Calotropis gigantea* were cut or scrapped freshly, washed with tap water, then with distilled water and then sun dried. The dried materials were powdered to a fine mesh of size < 75 µ and activated at 105°C for 4 hrs. in an oven and then these were employed in this work.

**(C): Adsorption experiment:**

**Batch system of extraction procedure was adopted** [1, 15, 36]. Carefully weighted quantities of adsorbents were taken into previously washed 1 lit/500 ml stopper bottles containing 500ml/250ml of Ammonium Chloride solution of predetermined concentrations. The various initial pH values of the suspensions were adjusted with dil. HCl or dil. NaOH solution using pH meter. The samples were shaken in mechanical shakers for a desired period and after the equilibration period; an aliquot of the sample was taken for Ammonia determination. Ammonia was determined by using Nessler’s method spectrophotometrically [37].

**(D) Estimation of Ammonia:**

An aliquot amount of ammonium chloride solution was taken in a 50ml volumetric flask. To it 1ml of Nessler’s reagent was added, mixed well and was allowed to stand at least 10min at room temp in a diffused light. The solution was diluted to the volume and mixed well. Optical Density of the orange-brown color was measured at 525 nm against a reagent blank using U.V and visible Spectrophotometer (of Systronics make). Thus obtained O.D Value was referred to standard graph (drawn between O.D and Concentration) prepared with known amounts of ammonia to find concentration of unknown solutions.

**(E) Effect of Interfering Ions:**

The interfering ions chosen for study are the common ions present in natural waters, viz., Sulphate, Nitrate, Chloride, Phosphate, Fluorides, Carbonate, Calcium, Magnesium, Copper, Zinc and Nickel. The synthetic mixtures of Chromium (VI) and one of the interfering ions were so made that the concentration of the interfering ions maintained at five fold excess than the ammonium ion concentration. 500 ml of these solutions were taken in stopped bottles and then
correctly weighed optimum quantities of the promising sorbents were added. Optimum pH was adjusted with dil. HCl or dil. NaOH using pH meter. The samples were shaken in shaking machines for the desired optimum periods and then the samples were filtered and analyzed for Ammonia. % of extraction was calculated from the data obtained. The results are presented in the Table No. 1.

(F) Applications of the developed bio-sorbents:
The adoptability of the methodology developed with the new bio-sorbents in this work for removing ammonia is tried with some real sewage/effluent samples of some industries and natural samples. For this purpose, samples were collected from the effluents of Paper pulp industry at Rajahmundry, Sugar industry at Tadepalligudem and Dairy form at Guntur in Andhra Pradesh. Further, three samples were collected at different polluted lakes in Bapatla mandalam of Guntur District of Andhra Pradesh.

Then these samples were subjected to extraction for Ammonia using the bio-sorbents developed in this work at optimum conditions of pH, equilibration time and sorbent concentration. The results obtained were presented in the Table 2.

RESULTS AND DISCUSSION

The leaf and stem/bark powders of Phyllanthus Niruri, Tridax procumbens, Morinda tinctoria, Azadirachta indica, Anonna squamosa and Calotropis gigantean have been found to have affinity towards ammonia. The percentage removal of ammonia is studied under various parameters viz., pH, time of equilibration and adsorbent dosage, with these bio-sorbents. The results obtained are presented in the Graph No: A: 1-6; B: 1&2; C: 1&2 and Table No. 1&2.
POWDER OF ANNONA SQUAMOSA LEAVES
Ammonia conc: 50ppm
Sorbent conc: 1.5g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: A-2a

POWDER OF STEM OF ANNONA SQUAMOSA
Ammonia conc: 50ppm
Sorbent conc: 3.0g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: 2b

POWDER OF CALOTROPIS ZYGANTIA LEAVES
Ammonia conc: 50ppm
Sorbent conc: 4.0g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: A-3.a

POWDER OF STEM OF CALOTROPIS ZYGANTIA
Ammonia conc: 50ppm
Sorbent conc: 4.0g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: 3b

POWDER OF TRIDOX PROCUMBENS LEAVES
Ammonia conc: 50ppm
Sorbent conc: 4.0g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: A-4.a

POWDER OF STEM OF TRIDOX PROCUMBENS
Ammonia conc: 50ppm
Sorbent conc: 3.0g/lit

Time in hours
Time vs% of removal of ammonia
Graph No: A-4.b
POWDER OF MORINGA TINCTORIA LEAVES
Ammonia conc:50ppm
Sorbent conc:4.0g/lit

POWDER OF BARK OF MORINGA TINCTORIA
Ammonia conc:50ppm
Sorbent conc:2.0g/lit

POWDER OF AZADIRACTA INDICA LEAVES
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

POWDER OF BARK OF AZADIRACTA INDICA
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

POWDER OF PHYLLANTHUS NERURU LEAVES
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

POWDER OF TRIDOX PROCUMBENS LEAVES
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

POWDER OF Moringa Tinctoria Leaves
Ammonia conc:50ppm
Sorbent conc:2.0g/lit

POWDER OF AZADIRACTA INDICA LEAVES
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

POWDER OF Calotropis Zygantia Leaves
Ammonia conc:50ppm
Sorbent conc:1.0g/lit

**Graph No:A-5.a**

**Graph No:A-5.b**

**Graph No:A-6.a**

**Graph No:A-6.b**

**Graph No:B:1**

**Graph No:B:2**

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The following observations are significant:

1. % of extractability increases with time for a fixed adsorbent at a fixed pH and after certain duration, the extractability remains constant, i.e. an equilibrium state has been reached (vide Graph Nos:A:1-6)

2. pH sensitivity: The % of extraction is found to be pH sensitive. The % of extractability of ammonia increases with time and decreases with the increase of pH for a fixed adsorbent concentration. (Vide Graph No. : A: 1-6; B: 1&2).

As for example, in the case of leaves powders of Phyllanthus nerui, the maximum extractability is found to be: 18.2% at pH: 10; 24.3% at pH: 9; 41.5% at pH: 8; 63.8% at pH: 6and 97% at pH: 5. In the case of leaves powders of Annona Squamosa, the maximum extractability is found to be : 21.8% at pH:10; 38.6% at pH:9; 48.3% at pH:8; 77.4% at pH:6and 95% at pH:5. With the leaves powders of Calotropis Zygantia, the extractability is found to be: 19.2% at pH: 10, 48.6 %at pH: 9; 59.1% at pH: 8; 68.3% at pH: 6and 85.0% at pH: 5. Tridox Procumbens leaves powders are extracted to an extent of: 25.7% at pH: 10; 30.2% at pH: 9; 42.8% at pH: 8; 63.8% at pH: 6and 88.0% at pH: 5. In the case of Moringa Tinctoria the extractability is found to be 19.8% at pH:10;37.3% at pH:9; 40.1% at pH:8 64.6% at pH:6 and 84.0% at pH:5. In the case of Azadiracta Indica the maximum extractability is found to be 21.8% at pH: 10; 37.4% at pH: 9; 58.2% at pH: 8; 76.7% at pH:6 and 95.0% at pH:5.

With the bark powders, the extractability at pHs: 10, 9, 8 ,7 ,6 and 5 is found respectively to be 20.8%, 29.3 %, 48.5 %, 73.8% and 100 % for Phyllanthus nerui; 25.2% 40.1%; 58.7%, 80.2% and 98.0% for Annona Squamosa; 26.3%,59.2 %; 67.5%; 78.8% and 91.0% for Calotropis Zygantia; 33.5%, 39.2%, 44.4%, 74.8% and 95.0% for Tridox Procumbens; 25.7%, 41.8%, 58.2%, 68.4% and 89.0% for Moringa Tinctoria; and 29.5%, 39.3 %, 62.8%,80.9% and 100.0% for Azadiracta Indica

3. The extraction of ammonia is found to be more in the case of bark powders as sorbents than powders of leaves. Further, the time needed for maximum removal of ammonia is also found to be less in the case of bark powders than powders of leaves. As for example , the maximum extractability of ammonia is 97% percent at pH: 5 after an equilibration period of 6 hrs. for Phyllanthus Neruri leaves powders while with its bark powders, the extractability is found to be found to be enhanced to 100% at pH:5 after an equilibration period of only 5 hrs. In the case of powders of leaves of Anona Squamosa, the % of extractability is found to be 95% at pH: 5 after an equilibration period of 6 hrs while with its bark powders the extraction increased to 98% at pH:5 after an equilibration period of only 5hrs. With leaves powders of Calotropis Zygantia, 85% of extraction of ammonia is found at pH:5 at an equilibration period of 6 hrs while with its bark powders, 91% of extraction is found at pH:5 and equilibration period of 5hrs is found to be adequate. Tridox Procumbens leaves powders are found to remove 88% ammonia at pH: 5 after an equilibration period of 6 hrs while with its bark powders, the % of removal is found to be 95% at pH:5 at equilibration period of 5hrs. Moringa Tinctoria leaves powders are found to remove 84% of ammonia at pH: 5 after an equilibration period of 6 hrs but its stems powders are found to remove 89% at pH: 5 after an equilibration period of 5hrs. In the case of Azadiracta Indica leaves powders, % of ammonia removal is found to be 95% at pH:5 after an equilibration
period of 6 hrs while with its bark ashes, 100% removal is observed after an equilibration periods of 6 hrs at pH:5.

4. When percentage removal is studied with respect to adsorbent dosage at fixed optimum pH: 5 and at optimum equilibration times, the graphs increase up to certain dosage and from then onwards plateaus are obtained vide Graph Nos. C:1 & 2).

With powders of leaves, the optimum sorbent dosage is found to be 1gm/lit for Azadiracta Indica, 1.5g/lit for Phyllanthus Neruri and Anona Squamosa, 4grms/lit for Calotropis Zygantia, Tridox Procumbens and Moringa Tinctoria (vide Graph Nos::C:1&2).

With the powders of barks, the sorbent concentration needed is considerably low. The sorption concentrations needed at optimum conditions of pH:5 and equilibration time are found to be is found to be 1 gm/lit for Phyllanthus Neruri, Anona Squamosa, and Azadiracta Indica, 2gms/lit for Moringa Tinctoria, 3 gms/lit for Tridox Procumbens and 3.5 gms/lit for Calotropis Zygantia (vide Graph Nos::C:1&2)

D: EFFECT OF INTERFERING IONS:
The extractions of Ammonia in presence of five fold excess of the common ions found in natural waters, namely Chloride, Fluoride, Sulphate, Phosphate, Carbonate, Calcium, Magnesium, Cupper and Zinc ions have been studied with the successful adsorbents at optimum conditions of pH:5 and time of equilibration and sorbent concentration as given in the Table 1. The results are presented in the Table 1.

The following observations are significant:
a. Cations namely, Ca^{2+}, Cu^{2+}, Zn^{2+} and Mg^{2+} ions are interfering with the % of extraction to some extent but in no case, % of extraction has come below 70.0% b. Anions of the present study viz., Chlorides, Fluorides, Sulphate, Phosphate and Carbonates are found to marginally affect the % of extractability of Ammonia under optimum experimental conditions.
Table No: 1: Effect of interfering Ions on the Extractability of Ammonia with different Bio-sorbents:

<table>
<thead>
<tr>
<th>S. No</th>
<th>Adsorbent and its concentration</th>
<th>Maximum Extractability at optimum conditions</th>
<th>% of Extraction of Ammonia in presence five fold excess of interfering ions at optimum conditions: Conc. of ammonia: 50 ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>SO$_4^{2-}$</td>
</tr>
<tr>
<td>1</td>
<td>Powder of Phyllanthus niruri leaves; 1.5 gms/lit</td>
<td>97.0%; pH:5, 6.0 hrs</td>
<td>92.0%</td>
</tr>
<tr>
<td>2</td>
<td>Powder Annona squamosa leaves; 1.5 gms/lit</td>
<td>95.0%; pH:5, 6.0 hrs</td>
<td>90.1%</td>
</tr>
<tr>
<td>3</td>
<td>Powder of Calotropis Zygantia leaves; 4.0 gms/lit</td>
<td>85.0%; pH:5, 6.0 hrs</td>
<td>83.2%</td>
</tr>
<tr>
<td>4</td>
<td>Powder of Tridax procumbens leaves; 0.0 gms/lit</td>
<td>88.0%; pH:5, 6.0 hrs</td>
<td>87.1%</td>
</tr>
<tr>
<td>5</td>
<td>Powder of Morinda tinctoria leaves; 4.0 gms/lit</td>
<td>84.0%; pH:5, 6.0 hrs</td>
<td>81.0%</td>
</tr>
<tr>
<td>6</td>
<td>Powder of Azadirachta indica leaves; 4.0 gms/lit</td>
<td>95.0%; pH:5, 6.0 hrs</td>
<td>92.1%</td>
</tr>
<tr>
<td>7</td>
<td>Powder of barks of Phyllanthus niruri 1.0 gm/lit</td>
<td>100.0%; pH:5.5, 5.0 hrs</td>
<td>95.1%</td>
</tr>
<tr>
<td>8</td>
<td>Powder of barks of Annona squamosa; 1.0 gm/lit</td>
<td>98.0%; pH:5, 5.0 hrs</td>
<td>94.2%</td>
</tr>
<tr>
<td>9</td>
<td>Powder of barks of Calotropis Zygantia; 3.5 gms/lit</td>
<td>91.0%; pH:5.5, 5.0 hrs</td>
<td>90.2%</td>
</tr>
<tr>
<td>10</td>
<td>Powder of barks of Tridax procumbens 3.0 gms/lit</td>
<td>93.0%; pH:5.5, 5.0 hrs</td>
<td>90.1%</td>
</tr>
<tr>
<td>11</td>
<td>Powder of Morinda tinctoria leaves; 2.0 gms/lit</td>
<td>89.0%; pH:5.5, 5.0 hrs</td>
<td>87.2%</td>
</tr>
<tr>
<td>12</td>
<td>Powder of Azadirachta indica leaves; 1.0 gm/lit</td>
<td>100.0%, pH:5.5, 5.0 hrs</td>
<td>95.0%</td>
</tr>
</tbody>
</table>
Table No.2: % of Extractability of Ammonia in Diverse Samples

<table>
<thead>
<tr>
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</thead>
<tbody>
<tr>
<td>Leaves powder of Phyllanthus Niruri at pH:5; Equilibration time: 6 hrs and sorbent concentration: 1.5 gms/litre</td>
<td>93.0 %</td>
<td>94.2%</td>
<td>90.2%</td>
<td>96.1%</td>
</tr>
<tr>
<td>Leaves powder of Annona squamosa at pH:5; Equilibration time: 6 hrs and sorbent concentration: 1.5 gms/litre</td>
<td>93.1%</td>
<td>91.8%</td>
<td>92.0%</td>
<td>91.1%</td>
</tr>
<tr>
<td>Leaves powder of Calotropis Zygantia at pH:5; Equilibration time: 6 hrs and sorbent concentration: 4.0 gms/litre</td>
<td>81.2%</td>
<td>82.1%</td>
<td>80.2%</td>
<td>83.0%</td>
</tr>
<tr>
<td>Leaves powder of Tridax procumbens at pH:5; Equilibration time: 6 hrs and sorbent concentration: 4.0 gms/litre</td>
<td>86.1%</td>
<td>87.1%</td>
<td>85.2%</td>
<td>86.5%</td>
</tr>
<tr>
<td>Leaves powder of Morinda tinctoria at pH:5; Equilibration time: 6 hrs and sorbent concentration: 4.0 gms/litre</td>
<td>82.0%</td>
<td>83.0%</td>
<td>81.5%</td>
<td>80.4%</td>
</tr>
<tr>
<td>Leaves powder of Azadirachta indica at pH:5; Equilibration time: 6 hrs and sorbent concentration: 4.0 gms/litre</td>
<td>91.1%</td>
<td>92.0%</td>
<td>90.2%</td>
<td>92.1%</td>
</tr>
<tr>
<td>Bark powder of Phyllanthus Niruri at pH:5; Equilibration time: 5 hrs and sorbent concentration: 1.0 gms/litre</td>
<td>94.5%</td>
<td>93.8%</td>
<td>94.2%</td>
<td>94.6%</td>
</tr>
<tr>
<td>Bark powder of Annona squamosa at pH:5; Equilibration time: 5 hrs and sorbent concentration: 1.0 gms/litre</td>
<td>93.2%</td>
<td>91.0%</td>
<td>90.2%</td>
<td>93.1%</td>
</tr>
<tr>
<td>Bark powder of Calotropis Zygantia at pH:5; Equilibration time: 5 hrs and sorbent concentration: 3.5 gms/litre</td>
<td>89.2%</td>
<td>88.2%</td>
<td>85.2%</td>
<td>89.1%</td>
</tr>
<tr>
<td>Bark powder of Tridax procumbens at pH:5; Equilibration time: 5 hrs and sorbent concentration: 3.0 gms/litre</td>
<td>93.1%</td>
<td>93.0%</td>
<td>94.1%</td>
<td>92.0%</td>
</tr>
<tr>
<td>Bark powder of Morinda tinctoria at pH:5; Equilibration time: 5 hrs and sorbent concentration: 2.0 gms/litre</td>
<td>86.2%</td>
<td>85.0%</td>
<td>83.0%</td>
<td>87.2%</td>
</tr>
<tr>
<td>Bark powder of Azadirachta indica at pH:5; Equilibration time: 5 hrs and sorbent concentration: 1.0 gms/litre</td>
<td>95.0%</td>
<td>96.5%</td>
<td>98.1%</td>
<td>98.0%</td>
</tr>
</tbody>
</table>
The scope of the present study does not permit to establish sound theoretical grounds for each observation made and infact, detailed investigations, are needed to account for the observations. However, the sorption characteristics of the adsorbents for ammonia may be generally viewed as follows:

- Ammonia exists in aqueous solution as either ammonium ion ($\text{NH}_4^+$) or ammonia gas depends upon the pH of the solution. Below pH: 9.25, the predominant species is $\text{NH}_4^+$ and above pH: 9.25, ammonia, $\text{NH}_3$, is considerable. In the present work, studies are made in the pH range from 5 to 10.

- Lingo celluloses like leaves and barks have weak ion affinity in the pH range of interest i.e. from 5 to 10. As pH decreases from 10 to 5, the equilibrium shifts towards the formation more and more $\text{NH}_4^+$ species, and thus formed positively charged species get exchanged to the sorbents and thereby progressively increasing the % of extraction. At high pH values, the predominant species is $\text{NH}_3$ and the species being neutral is uninfluenced by the electrostatic thrusts prevailing on the surface of the sorbent and hence, % of extraction decreases.

Applications:
The Applicability of the methodologies developed in this work have been tested with respects to the real samples of diverse nature, collected from the sewages/effluents of Paper pulp industry, Sugar Factory and also in natural polluted lakes. The results have been presented in the Table No: 2.

It is inferred from the Table No. 2 that the sorbents developed in this work are successful in removing Ammonia at optimum conditions of pH, equilibration time and sorbent dosage. % removal of Ammonia is found to be: 90.2% to 96.1% with leaves powder of Phyllanthus Niruri and 92.2% to 94.5% with their ashes ; 90.0% to 93.1% with leaves powder of Annona squamosa and 90.2% to 93.8 % with their ashes; 85.2% to 87.1% with the leaves powder of Tridax procumbens and 92.0% to 94.1% with their ashes; 80.4% to 83.2% with the leaves powder of Morinda tinctoria and 83.0 % to 87.4% with their ashes; 92.2% to 93.0 % with leaves powder of Azadirachta indica and 95.0% to 99.1% with their ashes; 80.2% to 84.0% with the leaves powder of Calotropis Zygantia and 85.2% to 89.2% with their ashes.

CONCLUSION

- The importance of bio-adsorbents such as leaves and barks in removing/controlling Ammonia from polluted waters is brought to the lime light.
- 97.0%, 95.0% , 85.0% , 88.0%, 84.0%, and 95.0% of removal ammonia are found with powders of leaves of Phyllanthus Niruri , Annona squamosa , Calotropis Zygantia , Tridax procumbens, Morinda tinctoria and Azadirachta indica respectively at pH:5 , equilibration time of 6.0 hrs and at optimum sorbent concentration.
- 100%, 98.0%, 91.0%. 95.0%, 89.0%and 100% of extractions of ammonia are observed for the powders of stems/barks of Phyllanthus Niruri , Annona squamosa , Calotropis Zygantia , Tridax procumbens, Morinda tinctoria and Azadirachta indica respectively at pH:5 at equilibration time of 5.0hrs and at optimum sorbent concentrations.
• The minimum sorbent dosage needed for the maximum removal of Ammonia is found considerably less for barks than for powders of leaves.

• **Interference:**
  Anions like Chlorides, Fluorides, Sulphate, Phosphate and Carbonates showed marginal interference while cations like $\text{Ca}^{2+}$, $\text{Cu}^{2+}$, $\text{Zn}^{2+}$ and $\text{Mg}^{2+}$ interfered to some extent. However, the % of extractability of Ammonia never comes down 70.0%.

• The methodologies developed in this work with the different bio-sorbents are found to be remarkably successful.

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**REFERENCES**