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Evaluating the Effectiveness of *Tamarindus Indica* Partially Activated Seed Coat Biomass in Removing of Nitrates from Aqueous Solutions

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ABSTRACT: Biomass derived from *Tamarindus indica* partially activated seed coat was investigated for the removal of nitrate ions from aqueous solutions. Batch experiments were performed to evaluate the parameters like pH, contact time, sorbent dose and initial nitrate concentration. pH of the solution played vital role. The maximum sorption observed at pH=7, sorbent dose 300mg, contact time at 120min, initial nitrate concentration 5mg. Physicochemical properties of the biomass were evaluated using scanning electron microscopy (SEM), energy dispersive X-ray analysis and Fourier Transform infra red (FTIR) spectroscopy. The SEM and FTIR data reveals the suitable surface and the presence of chemical functional groups such as hydroxyl, amide, carbonyl strong acid and primary amine on the biosorbent surface contributes to biosorption. The equilibrium isotherms and kinetics were deliberated. Biosorption equilibrium followed Langmuir isotherm. Pseudo second order kinetics provided better correlation of the experimental data in comparison with pseudo-first-order kinetic model. The study indicated that *Tamarindus indica* partially activated seed coat biomass found to be a novel biosorbent for the removal of nitrates from aqueous solutions.

Keywords: Biosorbent dose, pH, contact time, kinetic models, Isotherm.

INTRODUCTION

Due to amplified water demand, water scarcity is going to get worse in the forthcoming years. Water scarcity is already a reality of life in arid and semiarid regions where agricultural, domestic and industrial water demands compete for this limited resource. Continuous population growing growth, standards, rapid industrialization, using of chemical fertilizers and urbanization have been discharging organic and inorganic pollutants into freshwater resources (Katz & Dosoretz, 2008). Today nitrate is a most important water pollutant in many areas in the world such as Saudi Arabia (Nabil Alaa El-Din et al., 1994), India (Shrimali & Singh, 2001), UK (Neal et al., 2006), North America (Power & Schepers, 1989), Morocco (Sadeq et al., 2008), Changshu in China (Xie et al., 2007) and Toyserkan in western Iran (Jalali, 2011).

The most important sources of nitrogen are from nitrogenous compounds produced by plant and animals or the mining of sodium nitrate for use in fertilizers, and the

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atmosphere. The most oxidized structure of nitrogen is nitrate (NO3⁻) (Nixon, 1992; Bailey-Watts et al., 1993). High nitrate concentration in drinking water may carry us a variety of health effects. For instance, infants less than six months fed with nitrate contaminated water may possibly have blue baby syndrome, and if untreated, may die (Mousavi et al., 2012; Ghafari et al., 2008). In order to protect public health from the adverse effects of high nitrate intake, World Health Organization (WHO) set the standard as 50 mg/L to standardize the nitrate concentration in drinking water. The conventional methods (coagulation, filtration, and chlorination) applied for water treatment is not useful for nitrate elimination from water (Knobeloch et al., 2000).

There are a variety of technologies involving in the removal of nitrates from water and waste water, out of them biosorption has attained kind attention and based on nitrate binding capacities of different biological materials. Biosorption can be defined as the capability of biological resources to build up pollutants from wastewater through metabolically mediated or physico-chemical pathways of uptake (Fourest & Roux, 1992). In industrial sector, Biosorption has different advantages over exclusive clean technologies which were used for the treatment of water and waste water. The major advantages of biosorption, includes reusability of biomaterial, low operating cost, and high efficiency of pollutant removal from dilute solution. There is no supplementary need of nutrient requirement (Jagruti et al., 2015). The process of biosorption involves a solid phase which is a sorbent or biosorbent / biological material and a liquid phase which is a solvent, normally water containing a dissolved species to be sorbed which is sorbate (nitrate ions). Due to higher attraction of the sorbent for the sorbate species the sorbate is attracted and

detached by different mechanisms. The process continues till equilibrium is established among the amount of solid bound sorbate species and its portion remaining in the solution. The degree of sorbent attraction for the sorbate determines its allocation between the solid and liquid phases (Nilanjana Das et al., 2008).

The removal of nitrate ions from ground waters using different biosorbents is always of great attention (Mustaqeem et al., 2014). Treatment of ground water with sorbents of biological source is simple, comparatively inexpensive and friendly to the environment. Biosorption proves efficient, adaptable, powerful, and cost effective in the removal of different pollutants from industrial effluents.

The main objectives of this research were to: (i) To identify the optimum conditions required for *Tamarindus indica* partially activated seed coat biomass for effective removal of nitrates from aqueous solutions through batch experiment. (ii) To study the morphology and chemical structure of biosorbent. (iii) To study the isotherm and kinetic models.

MATERIAL AND METHODS

Tamarind seeds were collected from nearby Tamarind tree with in Venkateswara University campus, Tirupati, and transferred to research laboratory, Department of Environmental Sciences. Seeds were washed with distilled water to remove the dust particles. After drying, seeds were kept at hot air oven for 2-3 hours under 150° C temperature. When the seed coat color changed from medium dark to dark, taken out from oven and coats were separated from seeds. Separated coats were crushed into powder (biomass) up to uniform size (as shown in figure no.1). Prepared biomass was preserved in air tight glass bottle to protect from moisture for further studies (Shahanaz Begum et al., 2015).



Fig.1. Preparation of biosorbent from Tamarindus indica seed coat

Stock Solution: Weigh exactly 1.631 g of potassium nitrate and dissolve in water to make exactly 1,000 ml. Working standard solutions are prepared from these standard solutions at appropriate volumes.

Batch experiment: Batch experiments conducted by mixing of the were Tamarindus indica partially activated seed coat biomass with nitrate synthetic solution at different experimental conditions. In all sets of experiments, fixed volume of nitrate solution in 50mL was stirred with preferred biosorbent dose (50 - 350 mg) for the period of two hours. Different conditions of pH (3-8), initial concentrations $(1 - 6 \mu g)$ / mL) and contact time (30 – 150 minutes) were evaluated during the study. In order to adjust pH of the medium, 0.1 N of HCl and NaOH was used. The solutions were separated from the biomass by filtration through whatmann 40 filter paper. The initial and final concentrations of the nitrate ions in the solution were measured using UV-Visible Spectrophoto meter.

Nitarte ion removal efficiency (%) = [(C_0 - C_e)/ C_0] × 100

where C₀: Initial nitrate ion concentration of test solution, mg/L, C_e: Final equilibrium concentration of test solution mg/L.

RESULTS AND DISCUSSION

To understand the surface morphology and chemical compounds present on biosorbent SEM and FTIR analysis was done. Obtained results discussed below:

Scanning Electron Microscopy (SEM) with coupled Energy Dispersive Spectroscopy (EDS): To understand the morphology of biomass particles was observed in Scanning Electron Microscopy coupled Energy Dispersive with Spectroscopy (EDS). SEM images of untreated sample (before biosorption) of Tamarindus indica partially activated seed coat biomass exhibit non adhesive and dull appearance (as shown in Figure no. 2 a). The sharp edges of the surface are clearly visible. The Scanning Electron Micrograph

shows the typical expected shapes. The biomass is rich in porous and micro porous particles which can forms obvious gaps. These gaps give suitable binding sites for nitrate ions (Ramanaiah et al., 2007). In concerned with the porosity and the gaps among the particles is confirmed that the selected biosorbent has an adequate morphology to sorb nitrate ions (Venkata Mohan et al., 2002).

After biosorption, surface of

Tamarindus indica partially activated seed coat biomass become adhesive in appearance as evident from Fig. 2b. Elemental composition of pure and nitrate loaded biomass analyzed by SEM/EDX is shown in Fig. 2c and d. The weight percent of nitrate-loaded biomass gives nitrate peak apart from C and O peaks. The presence of prominent nitrate peak indicates that nitrate is superficially biosorbed on the surface of Tamarindus indica seed coat biomass.

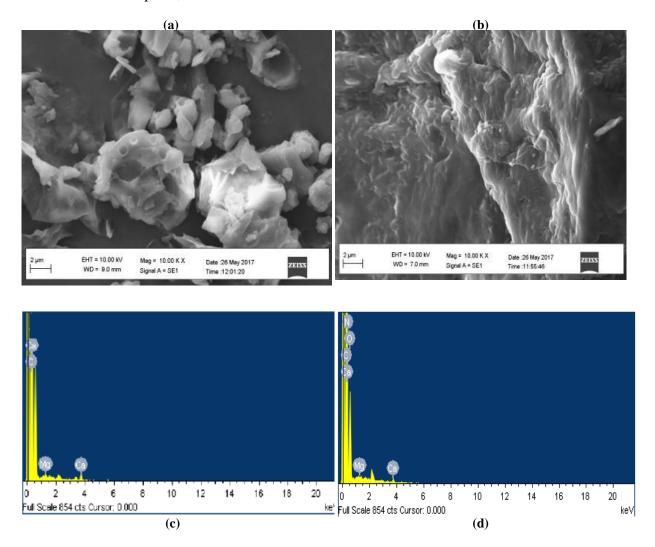


Fig. 2. Scanning Electron Microscopy (SEM) images of *Tamarindus indica* seed coat biomass. (a) pure (b) nitrate loaded and EDX images of *Tamarindis indica* seed coat (c) pure and (d) nitrate loaded.

FTIR (Fourier transform infrared) Spectroscopy: FTIR spectra of *Tamarindus indica* partially activated seed coat biomass after biosorption of nitrate (shown in figure no.3) was obtained to understand the nature

of the functional groups present in it. FTIR spectra displayed from 4000-500/cm many peaks which indicate the complex nature of biosorbent. The peak at 3417.88/cm indicates stretching of primary amine, the peak at

2923.41/cm due to stretching of O-H carbonyl strong acid, the peak 1620.36/cm is due to presence of amide (O=C-NH₂)group. The major differences in the region 1600-500/cm range is assigned to –NH₂ groups in different parts of the biomass. These stretching in functional groups are responsible for nitrate biosorption.

Effect of pH: In biosorption method hydrogen ion concentration is considered as one of the most significant parameters that influence the biosorption behavior of pollutant ions in aqueous solution (Yadav et al., 2013). It affects the solubility of the pollutant ions in the solution, replaces

some of the positive ions found in the active sites, and affects the degree of ionization of the sorbate during the reaction. The maximum biosorption was observed at pH 7.0 for nitrate ions (as shown in figure no.4) (Soumya Bikash, 2016). Only neutral pH was favorable acidic and basic conditions were not for maximum biosorption. favorable Therefore, the remaining all biosorption experiments were carried out at this pH value. The biosorption mechanisms on the Tamarindus indica seed coat surface reflect the character of the physicochemical interaction of the solution.

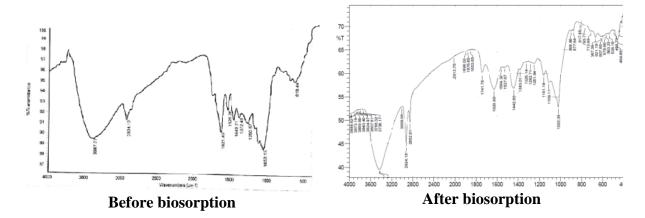


Fig. 3. FTIR Spectrum of *Tamarindus indica* partially activated seed coat biomass before and after nitrate biosorption.

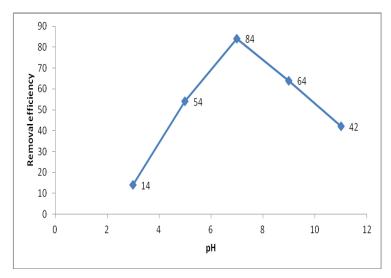


Fig. 4. Effect of pH on the biosorption

Biosorbent dose: Biosorbent (mg/ml) is a key parameter due to its effect on efficiency and on the amount of nitrate removed per unit weight of biomass. The effect of Tamarindus indica partially activated seed coat biosorbent dosage on the percentage removal of nitrate was studied. At first it was fluctuated and then increased with the increasing amount of the biosorbent as higher doses (as shown in figure no.5) of the biosorbent provided additional active sites and surface area (Bhaumik et al. 2012a, b). The maximum biosorption (84%) found to be at the dose of 300mg/50ml. The results can be attributed to the fact that increasing the biomass dosage gradually increases the biosorption sites for the nitrate ions (Mohammad Amer et al., 2015). Further increase of biosorbent dose has no significant increase in biosorption. situation which could be attributed to overlapping of biosorption sites as a result of overcrowding of biomass dose (Kalyan et al., 2013).

Effect of contact time: The rate of biosorption is important for designing batch biosorption experiments. Therefore, the effect of contact time on biosorption of nitrate ions by Tamarindus indica seed coat was investigated. All parameters such as dose of biosorbent and pH of solution were kept constant. biosorption of nitrate ions increased considerably until the contact time 120 minutes (Mohammad Amer et al., 2015). The results indicated that removal of nitrate was increased from 36 to 83 per cent with the contact time variation from 30 to 120 minutes. Thus the results illustrated that the optimum contact time for maximum removal of nitrate (83%) is 120 minutes (as shown figure no.6). This result is important because equilibrium time is one of the important parameters for an economical water treatment system.

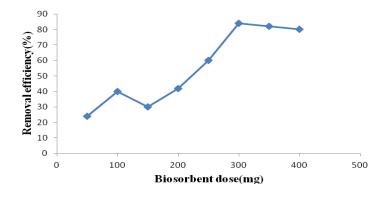


Fig. 5. Effect of biosorbent dose

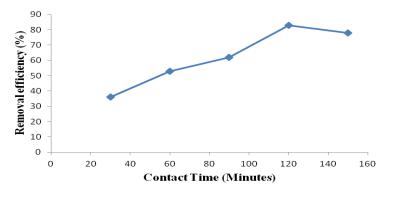


Fig. 6. Effect of contact time

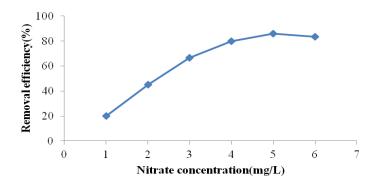


Fig. 7. Effect of initial nitrate concentration

Initial Nitrate concentration: The effect of initial nitrate ion concentration on biosorption rate was studied in the range (1-6 mg/L) at constant pH, and contact time. It was observed that the percentage removal decreased with increasing in initial nitrate concentration (Shown in figure no.7). The poorer uptake at higher nitrate concentration was resulted due to the increased ratio of initial number of moles of nitrate to the vacant sites available (Figucira et al., 2000; Hamdi et al., 2013). For a given biosorbent dose the total number of vacant sites available was fixed thus adsorbing almost the equal amount of adsorbate, which resulting in a decrease in the removal of adsorbate, consequent to an increase in initial nitrate concentration (Chu, 2002). Therefore it was evident from the results that nitrate biosorption was dependent on the initial nitrate concentration.

Biosorption isotherms: Biosorption isotherm describes the fraction of sorbate molecules that are partitioned among liquid and solid phases at equilibrium (Athar et al., 2014). Biosorption of nitrate ions onto *Tamarindus indica* partially activated seed coat was modeled using Langmuir, and Freundlich isotherms.

Langmuir isotherm model: The Langmuir isotherm assumes monolayer sorption on a homogeneous surface with a limited number of sorption sites. Once a site is filled, no more sorption can take place at that site. As such the surface will finally reach a saturation point where the maximum sorption of the surface will be achieved (Langmuir, 1918). The linear form of the Langmuir isotherm model is described as:

$$Ce/qe=1/K_L q_{max} + 1/q_{max} Ce$$

where, K_L is the Langmuir constant related to the energy of sorption and q_{max} is the maximum sorption capacity (mg/g). Values of Langmuir parameters, q_{max} and K_L were calculated from the slope and intercept of the linear plot of Ce/qe versus Ce as shown in Figure no. 8. Values of qmax, KL and regression coefficient R² are listed in Table 1. These values for Tamarindus indica seed coat biosorbent indicated that Langmuir describes the biosorption model phenomena as favorable. The essential characteristics of the Langmuir isotherm parameters can be used to expect the attraction between the sorbate and sorbent using separation factor or dimensionless equilibrium parameter, R_L, expressed as in the following Equation:

$$R_L = \frac{1}{1 + KL C0}$$

where, K_L is the Langmuir constant and Co is the maximum initial concentration of nitrate. The value of separation parameter R_L provides main information about the nature of sorption. The value of R_L indicated the type of Langmuir isotherm to be irreversible $(R_L=0)$, favorable $(0 < R_L < 1)$, linear $(R_L=1)$ or unfavorable $(R_L>1)$. The R_L for nitrate biosorption by *Tamarindus indica* partially

activated seed coat was found to be 0.63. They are in the range of 0–1 which indicates the favorable biosorption.

Freundlich isotherm: Freundlich isotherm model is the famous earliest relationship describing the sorption process. This model applies to sorption on heterogeneous surfaces with the interaction between sorbed molecules application of the Freundlich equation also suggests that sorption energy exponentially decreases on end of the sorption centers of sorbent. This isotherm is an empirical equation and can be employed to describe heterogeneous systems and is expressed as follows in linear form (Freundlich & Hellen, 1939):

$$\ln Q_e = \ln K_f + \frac{1}{n} \ln C$$

where, K_f is the Freundlich constant connected to the bonding energy. 1/n is the heterogeneity factor and n (g/L) is a measure of the deviation from linearity of sorption. Freundlich equilibrium constants were determined from the plot of log ge versus log Ce, Figure no.9. The n value indicates the degree of non-linearity among solution concentration and sorption as follows: if n = 1, then sorption is linear; if n<1, then sorption is a chemical process; if n>1, then sorption is a physical process. The n value in Freundlich equation was found to be 0.923 (Table no.1). Since n lie between 1 and 10, this indicates the physical sorption of nitrate ions onto Tamarindus indica seed coat. The values of regression coefficients R² are regarded as a measure of goodness of fit of the experimental data to the isotherm models.

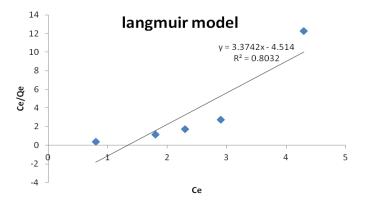


Fig. 8. Langmuir isotherm model

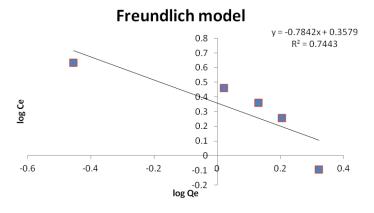


Fig. 9. Freundlich isotherm model

Biosorbent	pollutant	Langmuir parameters			Freundlich parameters		
		$\mathbf{Q}_{\mathbf{max}}$	$\mathbf{K}_{\mathbf{L}}$	\mathbb{R}^2	n	$\mathbf{K_f}$	\mathbb{R}^2
Tamarind seed coat	Nitrate	3.37	0.15	0.80397	0.923	0.86	0.744

Kinetic models: Kinetics of nitrate ion biosorption governs the rate determines the residence time and effectiveness of a biosorbent. In order to examine the mechanism of biosorption process, the two main types of biosorption kinetic models, namely reaction based (pseudo-first and pseudo-second order model) models were adopted to fit the experimental data.

Pseudo first order& second order: The pseudo-first order rate equation is given as:

$$\log (qe-qt) = \log qe - k1t/2.303$$

where, qt and qe are the amount of nitrate (mg/g) sorbed at time't' and equilibrium respectively, k1 is the rate constant of the pseudo-first order biosorption process (/min). Straight line plots of ln (qe-qt) against t were used to find out the rate constant k1, and sorption capacity.

The pseudo-second order equation is expressed as:

$$t/qt = 1/k_2 q_e^2 - (1/q_e)t$$

The plot of (t / qt) vs. t should give a linear relationship from which the values of the k_2 and qe could be determined from the intercept and slope of the plot, respectively.

From results it was concluded that biosorption of nitrate on *Tamarindus indica* seed coat biomass follow pseudo second order kinetics and concerned plots shown in figure no.10.

In present study, the linearized form of kinetics models and regression coefficient values indicates biosorption kinetics well fitted with pseudo-second-order kinetics. However, pseudo-first-order models showed moderate agreement with the studied results. The kinetics of sorbate and biosorbent interaction depends on many factors such as nature of sorbate and biosorbent, pH of the aqueous medium, temperature of the biosorption reaction, average time of contact between sorbate and biosorbent and mass transport process.

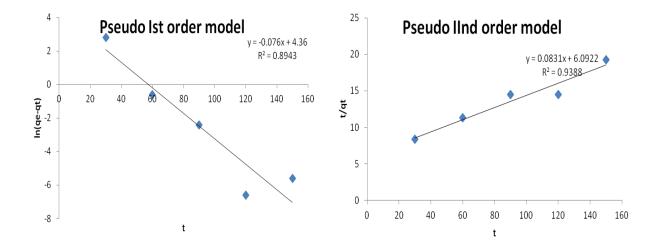


Fig. 10. Pseudo first and second order kinetic models

CONCLUSION

In the present research work, biosorption of nitrate from aqueous solutions using Tamarindus indica partially activated seed coat was drawn following conclusions. Tamarindus indica partially activated seed coat biomass has significant potential in the removal of nitrate from aqueous solutions. The SEM and FTIR data reveals the suitable surface and the presence of chemical functional groups such hydroxyl, amide, carbonyl strong acid and primary amine on the biosorbent surface contributes to biosorption. pH of the media has significant effect on nitrate removal and is best at pH 7. The optimum time for maximum biosorption was 120 minutes at the biosorbent dose of 300mg. Quantitative nitrate removal from water confirms the validity of these obtained results and the biosorption data for nitrate onto biomass were better correlated to the Langmuir and Freundlich isotherms. In the biosorption kinetic modeling studies, the pseudo second order chemical reaction kinetics provided the best correlation of the experimental data. The selected Tamarindus indica partially activated seed coat biomass was an effective, novel and eco-friendly biosorbent for the removal of nitrate from aqueous solutions.

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REFERENCES

Athar, M., Farooq, U., Ali, S.Z. and Salman, M. (2014). Insight into the binding of copper (II) by non-toxic biodegradable material (*Oryza sativa*): effect of modification and interfering ions. Clean Technol. Environ. Policy., 16; 579–590.

Bailey-Watts, A.E., Gunn, I.M.D. and Kirika, A. (1993). Loch Leven: Past and Current Water Quality and Options for Change. Edinburgh: Report to the Fourth River Purification Board, Institute of Freshwater Ecology

Bhaumik, R., Mondal, N.K., Das, B., Roy, P., Pal, K.C., Das, C., Banerjee, A. and Dutta, J.K. (2012a). Eggshell powder as biosorbent for removal of fluoride from aqueous solution: equilibrium, kinetic and thermodynamic studies. E. J. Chem., 9(3); 1457–1480.

Bhaumik, R., Mondal, N.K., Das, B., Roy, P., Pal, K.C., Das, C., Banerjee, A. and Dutta, J.K. (2012b). Eggshell powder as an adsorbent for removal of fluoride from aqueous solution: equilibrium, kinetic and thermodynamic studies. E. J. Chem., 9(3); 1457–1480.

Chu, K.H. (2002). Removal of copper from aqueous solution by chitosan in prawn shell: biosorption equilibrium and kinetics. J. Haz. Mat., 90(1); 77–95

Figucira, M.M.F., Volesky, B. and Cininelli, V.S.T. (2000). Biosorption of metals in brown sea wood biomass. Water Res., 34: 196–204.

Fourest, E. and Roux, J. C. (1992). Heavy metal biosorption by fungal mycelial by-products: mechanisms and influence of pH. Appl. Microbiol. and Biotech., 37; 399-403.

Freundlich, H.M.F. and Hellen, W. (1939). The adsorption of Cis- and Trans-Azobenzene. J. Am. Chem. Soc., 61; 2228–2230.

Ghafari, S., Hasan, M. and Aroua, M.K. (2008). Bio-electrochemical removal of nitrate from water and wastewater-a review. Bioresour. Technol., 99; 3965-3974.

Hamdi, W., Gamaoun, F., Pelster, D.E. and Seffen, M. (2013). Nitrate sorption in an agricultural soil profile. Appl. and Environ. Soil Sci., 4(3); 91-98.

Jagruti, N., Sandip, D., Maind. and Satish, A. (2015). Biosorption of Lead (II) and Chromium (VI) Onto Tarminalia Catappa L. Leaves: A Comparative Evaluation. J. Appli. Chem., 4 (6); 1700-1715.

Jalali, M. (2011). Nitrate pollution of groundwater in Toyserkan, western Iran. Environ. Earth Sci., 62; 907–913.

Kalyan, Y., Mok-Ryun Yu., Hoon Roh., Jae-Kyu Yang. and Yoon-Young Chang. (2013). Buffalo weed (Ambrosia trifida L. var. trifida) biochar for cadmium (II) and lead (II) adsorption in single and mixed system. Desalination and Water Treatment., 51:40-42; 7732-7745.

Katz. I. and Dosoretz, C.G. (2008). Desalination of domestic wastewater effluents: phosphate removal as pretreatment. Desalination., 222; 230–242.

Knobeloch, L., Salna, B., Hogan, A., Postle, J. and Anderson, H. (2000). Blue Babies and Nitrate

Contaminated Well Water. Environmental Health Perspectives Environ. Health Perspect., 108; 675-682.

Langmuir, I. (1918). Adsorption of gases on plain surfaces of glass, mica and platinum. J. Am. Chem. Soc., 40; 1361–1403.

Mohammad, W., Amer Rafat, A., Ahmad, A.M. and Awwad, M. (2015). Biosorption of Cu(II), Ni(II), Zn(II) and Pb(II) ions from aqueous solution by Sophora japonica pods powder. Int. J. Ind. Chem., 6; 67–75.

Mousavi, S., Ibrahim, M.K., Aroua, S. and Ghafari, S. (2012). Development of nitrate elimination by autohydrogenotrophic bacteria in bioelectrochemical reactors—A review. Biochem. Eng. J., 67; 251-264.

Mustaquem, M., Sharif Bagwan, M. and Patil, P. R. (2014). Removal of Pb (II) ion from aqueous Solution Using Potential Low Cost Adsorbents: An Isothermal modeling study. J. Appli. Chem., 3 (1): 275-282.

Nabil Alaa El-Din, M., Madany, I.M., Al-Tayaran, A., Hakeem Al-Jubair, A. and Gomaa, A. (1994). Trends in water quality of some wells in Saudi Arabia, 1984–1989. Sci. Total Environ., 143; 173–181.

Neal, C., Jarvie, H.P., Neal, M., Hill, L. and Wickham, H. (2006). Nitrate concentrations in river waters of the upper Thames and its tributaries. Sci. Total Environ., 365; 15–32.

Nilanjana Das, R., Vimala, K. and Karthika, P. (2008). Biosorption of heavy metals: an overview. Ind. J. Biotech., 7; 159-169.

Nixon, N. (1992). English water utility tackles nitrate removal. Water Eng. Manage., 139(3); 27–28.

Power, J.F. and Schepers, J.S. (1989). Nitrate contamination of groundwater in North America. Agr. Ecosyst. Environ., 26; 165–187.

Sadeq, M., Moe, C.L., Attarassi, B., Cherkaoui, I. and Idrissi, L. (2008). Drinking water nitrate and prevalence of methemoglobinemia among infants and children aged 1–7 years in Moroccan areas. Int J. Hyg. Environ. Health., 211; 546–554.

Shahanaz Begum, S.A., Tharakeswar, Y., Kalyan, Y. and Ramakrishna Naidu, G. (2015). Biosorption of Cd (II), Cr (VI) & Pb (II) from Aqueous Solution Using *Mirabilis jalapa* as Adsorbent. J. Encap. and Ads. Sci., 5; 93-104.

Shrimali, M. and Singh, K.P. (2001). New methods of nitrate removal from water. Environ. Pollut., 112; 351–359.

Soumya Bikash, G. (2016). Optimization study of adsorption parameters for removal of fluoride using aluminium-impregnated potato plant ash by response surface methodology. Clean Techn. Environ. Policy., 18; 1069–1083.

Ramanaiah, S.V., Venkata Mohan, S. and Sarma, P.N. (2007). Adsorptive removal of fluoride from aqueous phase using waste fungus (Pleurotus ostreatus 1804) biosorbent: Kinetics evaluation. Ecol. Eng., 3 (1); 47–56.

Venkata Mohan, S., Chandrasekhar Rao, N. and Karthikeyan, J. (2002). Adsorption removal of direct azo dye aqueous phase onto coal based sorbents: a kinetic and mechanistic study. J. Hazard. Mater., 90 (2); 189–204.

Xie, Y-X., Xiong, Z-Q., Xing, G-X., Sun, G-Q. and Zhu, Z-L. (2007). Assessment of nitrogen pollutant sources in surface waters of Taihu lake region. Pedosphere., 17; 200–208.

Yadav, A.K., Abbassi., R., Gupta, A. and Dadashzadeh, M. (2013). Removal of Fluoride from aqueous solution and groundwater by wheat straw, saw dust and activated carbon of sugarcane. Ecol. Eng., 52; 211–218.

