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The Potential Use of Natural Coagulants and Flocculants in the Treatment of Urban Waters

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Increasing population living in cities brings upon new challenges in water and wastewater management. It is therefore critical to improve the quality of urban water in a cost-effective and fossil-fuel approach in order to achieve a more sustainable future. Proper management of the natural water resources and its discharge back into the environment after use are of utmost importance due to its huge implication towards human developments. Hence, there have been ongoing investigations over the years in order to broaden the variety of methods for the treatment of urban water. Typical treatment processes to produce water safe for human consumptions include coagulation-flocculation, sedimentation, filtration and disinfection. In particularly, coagulation-flocculation process has always been a vital step to remove particulates, natural organic matters, microorganisms, inorganic ions, metals and others, thus significantly improves water quality. Increasing awareness of the health issues and environmental drawbacks regarding the use of conventional aluminium-based and iron-based coagulants shifted the interests towards plant- or animal-derived materials to be used as coagulants/flocculants. Unlike its inorganic counterparts, natural materials such as Moringa oleifera, tannin, chitosan, and seed gums were found to be effective, biodegradable, non-toxic to living organisms, and more environmentally friendly. This paper reviews the potential use of natural coagulants and flocculants in the treatment of urban waters for sustainable applications and consumptions in the cities.

1. Introduction

Water scarcity has become a global issue and is not only a problem limited to arid zones (Wu et al., 2013). Currently, many countries are confronted with significant water shortage due to the climate change, increasing agricultural and industrial activities and/or insufficient clean water supplies. It is predicted that the water shortage problem may worsen in the near future, judging from the escalating world's population and the anticipated increase in the water consumption per capita (Kalavrouziotis and Arslan-Alaton, 2008). As of 2008, the majority of the world populations live in the cities, leading to more challenges in water and wastewater managements in urban areas (Smith, 2009). It is believed that by 2020, more than half of the world's population will reside in urban areas (Aryal et al., 2010). Increasing environmental awareness has led to industries and businesses to constantly assess the impact of their activities on the environment and its contribution on resource depletion (Nouri et al., 2012). In this context, improving urban water quantity and quality in a cost effective manner was deemed to be a critical step towards achieving a sustainable future. According to Nasrabadi and Abbasi Maedeh (2014), groundwater quality is determined by geopogenic source (water-rock interaction), human intervention and climatic conditions. Although both surface water and groundwater provide a continuous mean of water supply, the presence of pathogenic microorganism, dissolved and suspended solids, colour and odour causing particles often renders the water to be unsafe for human activities (Choy et al., 2013). Wakida et al. (2014) suggested that to effectively remove turbidity, chemical oxygen demand (COD) and total phosphorus from the surface water originated from stormwater, the treatment facilities may have to remove particulates down to 10 μm. In developed countries, public institutions usually determine water quality objectives by considering health risks and encouraging appropriate treatment process to achieve these goals (Wu et al., 2013). Thus,

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proper water sanitations are often required to minimize pollutant load to an acceptable level before use. A typical urban water cycle without water reclamation in one city is depicted in Figure 1 (Amores et al., 2013).

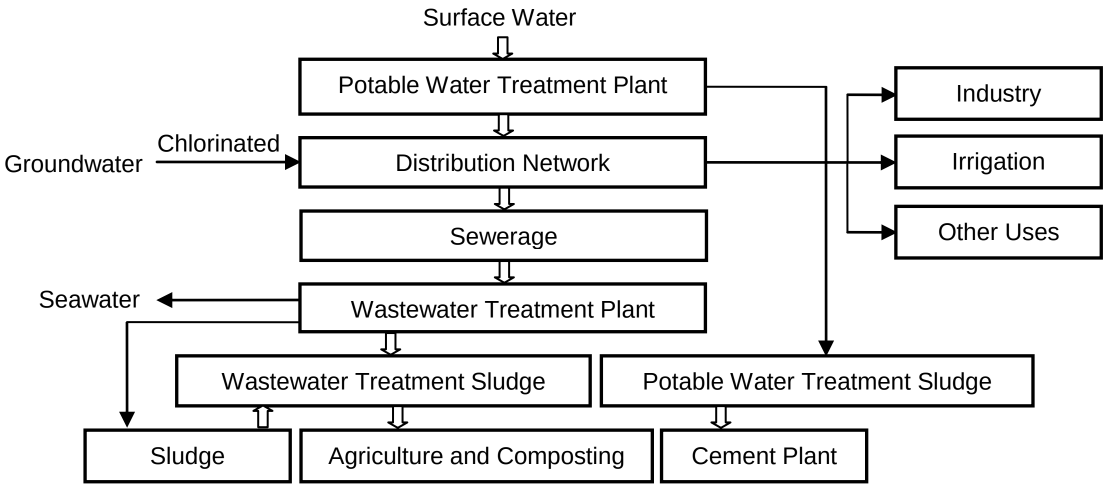

Figure 1: Typical urban water cycle (edited from Amores et al., 2013)

Figure 1 shows that potable water and wastewater treatment plants are usually involved in urban water cycle. Generally, both treatment plants involve physicochemical processes. Drinking would be the first and most important use for a metropolis (Nasrabadi and Abbasi Maedeh, 2014). Thus, drinking water requires meeting specific health requirements as the quality of the water is more crucial. Treatments in potable water treatment plants usually start with physicochemical treatments to separate the suspended particles and remove turbidity, following by chemical disinfection process (Amores et al., 2013). Although biological treatment is the main treatment process in wastewater treatment plants, physicochemical treatments are usually employed either as primary or tertiary treatment to remove particulate matters or to further improve the effluent quality before it is discharged into watercourses. Coagulation-flocculation process could be considered as one of the most typical physicochemical processes used in urban water treatments due to its easy operation, relatively simple design and low energy consumption. This process involves the introduction of coagulant or flocculant into the urban waters to agglomerate fine colloidal pollutant particles into large settleable flocs for its removal. Besides problem regarding sludge handling (Palahouane et al., 2013), an addition of inorganic coagulants in urban water may result in an increase of metal concentration in treated water supply and potentially contribute to Alzhelmer's disease (Teh et al., 2014). The use of coagulants/flocculants derived from plants or animals can potentially mitigate such complications. Generally, aggregation of the particles can occur via (a) double layer compression, (b) sweep flocculation, (c) adsorption and charge neutralization, and (d) adsorption and interparticle bridging. In particularly, the latter two mechanisms are closely associated with polymeric coagulants/flocculants derived from plants due to its long-chained structure which increases the number of unoccupied sites. Existence of background electrolytes in aqueous medium can facilitate the coagulating effect of natural polymeric coagulants as there is lower electrostatic repulsion between the particles (Yin, 2010). With various studies demonstrated the potential of using natural coagulants/flocculants in water management, this mini review aimed to summarize some of the important research related to the use of these materials for the treatment of urban waters.

2. Potential use of natural coagulants and flocculants in urban water treatment

The idea of utilizing natural coagulants and flocculants for water clarification dates back to over several millennia ago, way before the advent of chemical coagulants (Choy et al., 2013). The uses of natural coagulants and flocculants represent a vital development towards sustainable environmental technology as these materials are cost effective, highly biodegradable and unlikely to produce treated water with extreme pH (Yin, 2010). Table 1 summarizes some of the recent studies on the use of natural coagulants and flocculants such as Moringa oleifera (M. Oleifera), chitosan, tannins and gum seeds in urban water treatment. The use of these materials usually achieved similar or better turbidity removal efficiencies for treatment of waters with low-to-medium turbidity range (50-100 NTU) as compared to the established chemical coagulants such as alum (Yin, 2010). For instance, the use of cactus as a natural macromolecular coagulant at its optimum dosage was found to be able to achieve similar turbidity removal as compared to alum or polyferric sulphate (PFS) in the treatment of sewage or potable water (Zhang et al., 2006). Natural coagulants could also be used to solve the high fluoride content in waters which could cause dental or skeletal fluorosis upon consumption (Dos Santos Bazanella et al., 2012). Spikes in fluoride levels in groundwaters are often caused by industrial production of phosphate fertilizers and volcanic activities. It was found that coagulation using M. oleifera could effectively remove up to 90 % of fluoride content of the treated water, making it safe for human consumption (Dos Santos Bazanella et al., 2012).

Performance of chitosan, a linear cationic polymer, also surpassed alum for the treatment of highly turbid raw water in terms of residual turbidity of treated water and the amount of sludge produced. Chitosan was able to effectively remove turbidity due to the interactions between its functional groups and the active sites of pollutant particles. Attachment of particles onto chitosan chain forms large aggregates via bridging mechanism which settles under the action of gravity (Hu et al., 2013). The use of natural coagulants is also advantageous as it does not significantly disrupt the pH of the treated water. In the treatment of well water, an addition of M. oleifera, guar gum or Jatropha curcas only caused a small scatter in pH values for dosages of 1 to 500 mg/L. The pH of treated water was well within the WHO (2006) recommendation of pH 6.5 to pH 8.5. In contrast, an addition of alum decreased the pH of water to pH 5.6 due to its acidic nature. Thus, treatment using alum requires extra cost for chemicals to raise the pH back to the allowable values (Pritchard et al., 2009).

Study showed that coagulant protein extracted from Mustard seed performed better than seed extracts from M. oleifera in the treatment of highly turbid pond water (Bodlund et al., 2013). Protein profile analysis on Mustard seed indicated the presence of coagulant protein with sizes of approximately 9 and 6.5 kDa, similar to those obtained from M. oleifera coagulant protein and napin3 protein, respectively (Bodlund et al., 2013). Derivatives of cellulose, the most abundant biopolymer on earth, were also been investigated to produce natural flocculants. For example, the performance of dicarboxylic acid nanocellulose (DCC) flocculants produced by nanofibrillation of periodate and chlorite-oxidized celluloses with a homogenizer (Suopajärvi et al., 2013). Cellulose raw material used for this synthesis was obtained from bleached birch (Betula verrucosa and B. pendula) chemical wood pulp. During the treatment of municipal wastewater, DCC with high charge density and high nanofibril content gave the best flocculation performance. In combination with ferric sulphate, DCC synthesized were able to achieve turbidity and COD removal similar to that of commercial reference flocculant (Fennopol K1360, cationic polyacrylamide) (Suopajärvi et al., 2013).

Some natural coagulants/flocculants have antimicrobial property which could aid in controlling pathogenic microorganism which causes infections and water-borne diseases in potable water production (Ahmed et al., 2010). The use of M. oleifera plant extract as a more sustainable and cost effective water purification technique is promising as it showed good turbidity with high removals of total coliforms and Escherichia coli in both reservoir waters and turbid rivers (Pritchard et al., 2010). It was reported that stagnant water treated with M. oleifera seeds has a total viable bacterial count of only 1,000 CFU/mL, a significantly lower value as compared to 9,000 CFU/mL obtained by using alum as a coagulant. Further investigation showed that the buds taken from M. oleifera have a great potential as antibacterial compounds against specific water-borne pathogens as compared to its shoot, leaves or seeds (Ahmed et al., 2010).

3. Possible improvements of coagulation-flocculation treatment of urban waters using natural coagulants and flocculants

3.1 Combination of natural and inorganic coagulants or flocculants

Combining both natural and inorganic coagulants or flocculants in water treatment is also promising. A study showed that the use of cactus with alum for sewage water treatment improved turbidity and COD removal as compared to the treatment using either coagulant alone (Zhang et al., 2006). The combined use of both M. oleifera (100 mg/L) and alum (10 mg/L) to treat sewage water also successfully achieved the best COD removal (64 %) in a combined treatment processes consisting of coagulation-flocculation, sedimentation and sand filtration (Bhuptawat et al., 2007). As for the treatment of highly turbid surface water, coagulation-flocculation using chitosan and a small dose of aluminium salt effectively solved the high residual turbidity of treated water which renders the feasibility of the subsequent sand filtration when chitosan was used alone. In addition, the amine groups in chitosan also helped lower the residual aluminium concentration by adsorbing the metal cations, thus reducing the risk of Alzheimer’s disease (Hu et al., 2013). Integrating coagulation-flocculation process using natural coagulant with other processes is also one of the techniques to effectively treat urban water. For example, Bergamasco et al. (2011)

combined coagulation step using chitosan with ultrafiltration for water disinfection. Although high membrane fouling was observed, this integrated treatment yielded higher stabilized permeate flux, which was twice of that achieved by using alum.

Table 1: Studies on the use of natural coagulants/flocculants for urban water treatment

Coagulant and flocculant	Wastewater	Removal Efficiencies (Dosage)	Remarks	Reference
Moringa oleifera	Raw surface water	Turbidity: ~75 % (0.5 mg/L); DOC: ~50 % (2.5 mg/L)	Two-step purification of *M. oleifera* had significantly better turbidity removal and lower residual DOC as compared to single-step purification.	Sánchez-Martín et al. (2010)
	Surface water	Turbidity: ~98 % (16 g/L); *E.coli*: ~50 % (16 g/L)	Filtration step was necessary to remove organic matter contributed by coagulant.	Poumaye et al. (2012)
	Shallow wells	Turbidity: 100 % (250 mg/L)	*M. oleifera* exhibited higher turbidity removal as compared to guar gum and *jatropha curcas*	Pritchard et al. (2011)
	Groundwater	Flouride: 90.9 % (2.5 g/L)	Interaction between *M. oleifera* and negative charge of fluoride occurred through ion adsorption mechanism. Ultrafiltration post-treatment was performed to remove colour and turbidity caused by coagulation.	Santos Bazanella et al. (2013)
Cactaceous optunia	Sewage water	Turbidity: ~87 % (60 mg/L); COD: ~55 % (50 mg/L)	Combined cactus and alum showed better efficiency as compared to cactus or alum alone.	Zhang et al. (2006)
	Potable water source	Turbidity: ~93 % (50 mg/L)	Turbidity removal using cactus was similar to alum and polyferric sulphate	Zhang et al. (2006)
Chitosan	Surface water	Turbidity: ~99.3 % (1 mg/L); COD: 59.9 % (1 mg/L)	Chitosan showed better COD and TDS removal as compared to alum	Bergamasco et al. (2011)
	Raw water	Turbidity: ~99 % (1-2 mg/L)	Amount of sludge produced using chitosan was significantly lesser as compared to alum and did not increase with increasing dosage of chitosan.	Hu et al. (2013)
	Surface water	Turbidity: ~75 % (1 mg/L)	Chitosan may result in more toxic finished water as compared to alum and ferric chloride	Rizzo et al. (2008)
Modified tannin	Municipal wastewater	Turbidity: ~100 % (100 mg/L)	Tannin extract from *Acacia mearnsii de Wild* was modified via cationisation process using ammonium chloride and formaldehyde. 30% surfactant removal was achieved.	Beltrán-Heredia et al. (2011)
	Surface water	Turbidity: ~100 % (25 mg/L)	Low level coagulants were able to remove large majority of natural turbidity in sample.	Beltrán-Heredia et al. (2011)
Modified guar gum	Municipal wastewater	TSS:~85 % (9 mg/L), COD: ~61 % (9 mg/L)	Polyacylamide grafted carboxymethyl guar gum (CMG-*g*-PAM) was synthesized using microwave assisted grafting method.	Pal et al. (2011)
Modified gum ghatti	Municipal wastewater	Final turbidity: 29 NTU (8 mg/L) Final COD: 24 mg/L (8 mg/L)	Polyacrylamide chains (PAM) were grafted onto backbone of gum ghatti by microwave assisted method.	Rani et al. (2012)

3.2 Modifications of natural coagulants or flocculants

Currently, there have been studies focussing towards improving the performance of natural plant or animal extracts to be used as a coagulant and flocculant through modifications. For example, gum ghatti, cheap and abundant polysaccharide, cultivated and exported from Indian subcontinent, was modified by grafting polyacrylamide chains (PAM) onto its backbone via microwave assisted method. The polyacrylamide grafted gum ghatti achieved 4 times higher turbidity removal in treatment of municipal wastewater as compared to unmodified gum ghatti (Rani et al., 2012). Polyacrylamide grafted carboxymethyl guar gum prepared by microwave assisted grafting method also exhibited better flocculating performance as compared to unmodified guar gum. Besides reducing the pollutant load (e.g. TS, TSS, TDS, turbidity, COD), various metals (e.g. chromium, nickel) present in the municipal wastewater was also effectively removed using this modified guar gum (Pal et al., 2011). Tannin obtained from Acacia mearnsii bark was also modified via physicochemical process to produce a flocculant with high flocculating properties. This modified flocculant presents a more environmentally friendly alternative for coagulation-flocculation treatment of raw water because its ability to remove BOD5, COD and turbidity were comparable to alum (Beltrán-Heredia and Sánchez-Martín, 2009).

A newly developed composite coagulant was obtained by hybridizing polyaluminium hydroxychloride (PACI) with chitosan (PACI-chitosan) for the treatment of natural water (Ng et al., 2013). Besides charge neutralization (due to the positive charges from Al^{3+} and amine bonds) and hydrogen bonding interaction, the presence of chitosan helps in polymeric bridging where the long chain polymers adsorb onto the surface of pollutant particles to form larger flocs. PACI-chitosan was more effective than PACI in treating water samples containing higher level of activated polyhydroxyaromatic moieties (Ng et al., 2013).

3.3 Purification of natural coagulants or flocculants

Although many advantages could be obtained through natural coagulants and flocculants, there are some challenges from its usage. Several studies reported an increase in residual dissolved organic carbon (DOC) (Sánchez-Martín et al., 2010) or increase in colour (Santos Bazanella et al., 2013) from coagulation using plant extract making its use in drinking water not feasible. Sánchez-Martín et al. (2010) suggested coagulant purification using ion-exchange processes which could significantly enhance its treatment performance while lowering the extent of residual DOC in treated water.

4. Conclusions

Increasing demand for eco-friendly technology has sparked the emergence of plant or animal derived alternatives as replacements for conventional metal salt coagulants in coagulation-flocculation for treatment of urban waters. Studies showed that these natural alternatives could perform better than or comparable to its inorganic counterpart with additional environmental benefits such as high biodegradability and good antimicrobial property. Despite its huge potential, the mechanisms associated with different natural coagulants/flocculants are often poorly understood, unlike the widely developed chemical coagulants. Attention should also be given towards investigating the various operating parameters (e.g. coagulant/flocculant dosage, initial pH, settling time, etc.) which could significantly affect the viability and effectiveness of these natural materials for the treatment of urban waters. Furthermore, issues such as absence of mass plantation of the plants, perceived low-volume market and non-existent supportive regulations for quality control of the produced coagulant extracts has to be addressed before introducing plant- or animal-derived coagulants/flocculants for large-scale water treatment purposes.

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References

Ahmed T., Kanwal R., Hassan H., Ayub N., Scholz M., McMinn W., 2010, Coagulation and disinfection in water treatment using Moringa, Proc. Inst. Civil. Eng.- Water Manag., 163, 381-388.

Amores M.J., Meneses M., Pasqualino J., Antón A., Castells F., 2013, Environmental assessment of urban water cycle on Mediterranean conditions by LCA approach, J. Clean. Prod., 43, 84-92.

Aryal R., Vigneswaran S., Kandasamy J., Naidu R., 2010, Urban stormwater quality and treatment, Korean J. Chem. Eng., 27, 1343-1359.

Beltrán-Heredia J., Sánchez-Martín J., 2009, Municipal wastewater treatment by modified tannin flocculant agent, Desalination 249, 353-358.

Beltrán-Heredia J., Sánchez-Martín J., Dávila-Acedo M.A., 2011, Optimization of the synthesis of a new coagulant from a tannin extract, J. Hazard. Mater., 186, 1704-1712.

Bergamasco R., Konradt-Moraes L.C., Vieira M.F., Fagundes-Klenb M.R., Vieira A.M.S., 2011, Performance of a coagulation-ultrafiltration hybrid process for water supply treatment, Chem. Eng. Res., 166, 483-489.

Bhuptawat H., Folkard G.K., Chaudhari S., 2007, Innovative physico-chemical treatment of wastewater incorporating Moringa oleifera seed coagulant, J. Hazard. Mater., 142, 477-482.

Bodlund I., Pavankumar A.R., Chelliah R., Kasi S., Sankaran K., Rajarao G.K., 2013, Coagulant proteins identified in Mustard: a potential water treatment agent, Int. J. Environ. Sci. Technol., 11, 873-880.

Choy S.Y., Prasad K.M.N., Wu T.Y., Ramanan R.N., 2013, A review on common vegetables and legumes as promising plant-based natural coagulants in water clarification, Int. J. Environ. Sci. Technol., DOI: 10.1007/s13762-013-0446-2

Dos Santos Bazanella G.C., da Silva G.F., Vieira A.M.S., Bergamasco R., 2012, Flouride removal from water using combined Moringa oleifera/ultrafiltration process, Water Air Soil Pollut., 223, 6083-6093.

Hu C.-Y., Lo S.-L., Chang C.-L., Chen F.-L., Wu Y.-D., Ma, J.l., 2013, Treatment of highly turbid water using chitosan and aluminum salts, Sep. Purif. Technol., 104, 322-326.

Kalavrouziotis I.K., Arslan-Alaton I., 2008, Reuse of urban wastewater and sewage sludge in the Mediterranean countries: Case studies from Greece and Turkey, Fresenius Environ. Bull., 17, 625-639.

Nasrabadi T., Abbasi Maedeh P., 2014, Groundwater quality degradation of urban areas (case study: Tehran city, Iran), Int. J. Environ. Sci. Technol., 11, 293-302.

Ng M., Liu S., Chow C.W.K., Drikas M., Amal R., Lim M., 2013, Understanding effects of water characteristics on natural organic matter treatability by PACl and a novel PACl-chitosan coagulants, J. Hazard. Mater., 263, 718-725.

Nouri J., Nouri N., Moeeni M., 2012, Development of industrial waste disposal scenarios using life-cycle assessment approach, Int. J. Environ. Sci. Technol., 9, 417-424.

Pal S., Ghorai S., Dash M.K., Ghosh G., Udayabhanu G., 2011, Flocculation properties of polyacrylamide grafted carboxymethyl guar gum (CMG-g-PAM) synthesized by conventional and microwave assisted method, J. Hazard. Mater., 192, 1580-1588.

Palahouane B., Drouiche N., Bensadok K, Aoudjm S., Hecini, M., 2013. Remediation of post treated fluorinated photovoltaic wastewater by electrocoagulation. Chem. Eng. Trans., 32, 25-30.

Poumaye N., Mabingui J., Lutgen P., Bigan M., 2012, Contribution to the clarification of surface water from the Moringa oleifera: Case M'Poko River to Bangui, Central African Republic, Chem. Eng. Res. Des., 90, 2346-2352.

Pritchard M., Craven T., Mkandawire T., Edmondson A.S., O'Neill J.G., 2010, A comparison between Moringa oleifera and chemical coagulants in the purification of drinking water – An alternative sustainable solution for developing countries, Phys. Chem. Earth, 35, 798-805.

Pritchard M., Mkandawire T., Edmondson A., O'Neill J.G., Kulilanga G., 2009, Potential of using plant extracts for purification of shallow well water in Malawi, Phys. Chem. Earth, 34, 799-805.

Rani P., Sen G., Mishra S., Jha U., 2012, Microwave assisted synthesis of polyacrylamide grafted gum ghatti and its application as flocculant, Carbohydrate Polym., 89, 275-281.

Rizzo L., Gennaro A.D., Gallo M., Belgiorno V., 2008, Coagulation/chlorination of surface water: A comparison between chitosan and metal salts, Sep. Purif. Technol., 62, 79-85.

Sánchez-Martín J., Ghebremichael K., Beltrán-Heredia J., 2010, Comparison of single-step and two-step purified coagulants from Moringa oleifera seed for turbidity and DOC removal, Bioresource Technol., 101, 6259-6261.

Santos Bazanella G.C., Silva G.F., Viera A.M.S., Bergamasco F., 2008, Flouride removal from water using combined Moringa oleifera/ultrafiltration process, Water Air Soil Pollut., 223, 6083-6093.

Smith B.R., 2009, Re-thinking wastewater landscapes: combining innovative strategies to address tomorrow's urban wastewater treatment challenges, Water Sci. Technol., 60, 1465-1473.

Suopajärvi T., Liimatainen H., Hormi O., Niinimäki J., 2013, Coagulation-flocculation treatment of municipal wastewater based on anionized nanocelluloses, Chem. Eng. J., 231, 59-67.

Teh C.Y., Wu T.Y., Juan J.C., 2014, Optimization of agro-industrial wastewater treatment using unmodified rice starch as a natural coagulant., Ind. Crop. Prod., 56, 17-26.

Wakida F.T., Martinez-Huato S., Garcia-Flores E., Piñon-Colin T.D.J., Espinoza-Gomez H., Ames-López A., 2014, Pollutant association with suspended solids in stormwater in Tijuana, Mexico, Int. J. Environ. Sci. Technol., 11, 319-326.

WHO (World Health Organisation), 2006, Guidelines for Drinking-Water Quality, First Addendum to Third Edition. Recommendations, Vol. 1., <www.who.int/water_sanitation_health/dwq/gdwq0506.pdf> accessed on 09.03.2014

Wu T.Y., Mohammad A.W., Lim S.L., Lim P.N., Hay J.X.W., 2013, Chapter 3: Recent Advances in the Reuse of Wastewaters for Promoting Sustainable Development, Wastewater Reuse and Management, Springer, the Netherlands, 47-103

Yin C.-Y., 2010, Emerging usage of plant-based coagulants for water and wastewater treatment, Process Biochem., 45, 1437-1444.

Zhang J., Zhang F., Luo Y., Yang H., 2006, A preliminary study on cactus as coagulant in water treatment, Process Biochem., 41, 730-733.