Constructed wetlands using aluminium-based drinking water treatment sludge as P-removing substrate: Should aluminium release be a concern?

Akintunde O. Babatunde[1], Jeyakumar L.G. Kumar and Yaqian Zhao*

**Centre for Water Resources Research, School of Architecture, Landscape and Civil Engineering, University College Dublin, Belfield, Dublin 4, Ireland.*

**Corresponding author: Tel: +353-1-7163215; Fax: +353-1-7163297;*
E-mail: yaqian.zhao@ucd.ie

[1]Present address: Discipline of Civil Engineering, School of Computing, Science and Engineering, University of Salford, Salford, M5 4WT, Greater Manchester, U.K.

Abstract

This study investigated an important issue of aluminium (Al) release in a novel reuse of Al-based water treatment sludge (Al-WTS) in constructed wetland system (CWs) as alternative substrate for wastewater treatment. Al-WTS is an inevitable by-product of drinking water treatment plants that use Al-salt as coagulant for raw water purification. It has recently been demonstrated that Al-WTS can be reused as a low-cost phosphorus (P) adsorbent and biofilm carrier in CWs for wastewater treatment. However, to facilitate the large scale application of Al-WTS in CWs as wetland substrate, concerns about Al leaching during its' reuse in CWs must be addressed as Al is a dominant constituent in Al-WTS. In this study, a desk review of literature on Al release during Al-WTS reuse was conducted. Furthermore, a 42-week Al monitoring was carried out on a pilot field-scale CWs employing Al-WTS as main substrate. Results show that 22 out of the 35 studies reviewed, reported Al release with levels of soluble Al reported ranging from 0.01 to about 20 mg L^{-1}. Monitoring of Al in the pilot field-scale CWs shows that there was Al leaching. However, except for the first three weeks of operation, effluents concentrations of both total- and soluble-Al were all below the general regulatory guideline limit of 0.2 mg L^{-1}. Overall, the study addresses a very vital concern regarding the successful application of Al-WTS in CWs and shows that Al release during such novel reuse is quite low and should not preclude its use.

Keywords: Aluminium, alum sludge, constructed wetlands, leaching, water treatment sludge, wastewater treatment

1. Introduction

Virtually all drinking water treatment facilities worldwide generate an enormous amount of water treatment sludge (WTS) for which environmentally friendly end-use options are continually being sought as opposed to landfilling. Babatunde and Zhao [1] identified eleven ways in which WTSs are currently being reused. However, one innovative option which holds great promise is the beneficial reuse of WTSs as phosphorus (P) adsorbent and as substrate in constructed wetland system (CWs) for wastewater treatment.[2] In particular, aluminium-based WTS (Al-WTS) has attracted considerable attention as it is the most widely generated WTS worldwide. It offers huge benefits particularly for P removal and biofilm attachment when used as substrate in CWs.[2]

The origin of the use of dewatered Al-WTS as main substrate in CWs lies in the fact that performance of CWs is good in terms of removal of organics and suspended solids (SS), but their performance as regards nutrient reduction (especially P) has been inconsistent and often low. Consequently, several alternative substrates with high P sorption capacity have been tested mostly in laboratory scale as potential adsorbents to reduce P concentration to acceptable levels in CWs. Investigations revealed that Al-WTS has suitable physicochemical characteristics for use as CWs substrate and it is mainly composed of amorphous aluminium which greatly enhances P adsorption capacity.[3] By reusing it as CWs substrate, P removal can be greatly enhanced while at the same time, a sustainable disposal alternative which converts Al-WTS from residual into a resource for wastewater treatment will have been developed. However, since Al is a dominant constituent of Al-WTS, there are concerns about Al leaching during Al-WTS reuse in CWs for wastewater treatment. Such concerns need to be addressed and potential risk to the environment must be properly evaluated before any large-scale application of Al-WTS in CWs can be assured.

There are numerous environmental sources of Al and it is commonly found in natural waters of pH values greater than 4.0 at concentrations less than 0.1 mg L^{-1}, and this is found to increase with pH decrease.[4] The toxicity of Al is a widespread problem in all forms of life, including humans, animals, fish and plants. Al is a neurotoxicant which has been shown to play a role in the etiology of uremia and dialysis-associated disorders of the brain (dialysis encephalopathy) and bone (aluminium-associated bone disease).[5] Al is also acutely toxic to fish in acid waters,[6] while it is also known to cause toxicity to aquatic habitat.[7] In plants, phytotoxic Al ion (mainly Al^{3+}) restrict crop productivity in acidic soils which cover almost 40% of world's arable land.[8] Mahdy et al. [9] noted that the toxic effects of Al on plants are observed in association with soluble Al (Al^{3+}) that is biologically available in acidic soil and water (pH < 5.5) but is biologically inactive in circumneutral to alkaline (pH 5.5–8.5) conditions.

Therefore, because of the general interest in aluminium toxicity as an environmental and health threat, it becomes very crucial to ascertain if Al is released during any reuse of Al-WTS and to determine if such release is within safe limits. This study is thus concerned with addressing and investigating such concerns by monitoring the release of Al when Al-WTS is used as substrate in a pilot field-scale CWs. A desk review was firstly carried out to inform of the current opinion and experiences on the release of Al when Al-WTS is reused for beneficial purposes.

2. Literature review on Al release from studies focusing on various Al-WTS reuses

A preliminary screening of published works on the various reuses of Al-WTS for P removal was conducted. Emphasis was placed on any data and/or information relating to the release of Al during such reuse. Literature searches were conducted using two main databases; the ISI web of science (with proceedings) and Scopus, while general searches were also conducted using web-search engines. The terms alum sludge reuse and reuse of aluminium-water treatment residual were used individually and in combinations with other terms such as phosphorus removal, aluminium release and aluminium leaching.

Table 1 shows the results of the desk review conducted with regard to the current state of knowledge on Al release during Al-WTS reuse particularly for P removal. A total of 35 related studies were reviewed. Except for the studies where Al release was not particularly monitored or studied, all the other studies reportedly observed Al release. Out of the 35 studies reviewed, Al release was mentioned or observed in 22 studies. However, although Al release was not reported in 13 of the studies, it does not suffice to conclude whether Al was released or not. Concentration of leached/soluble Al reported ranged from 0.01 to about 20 mg L^{-1} (Fig. 1) and the influence of pH was mostly noted. However, except for three reported values of 1.11 mg L^{-1}, 7 mg L^{-1} (pH 3) and about 20 mg L^{-1} (pH 3), all other values reported were < 1 mg L^{-1} and mostly between <0.01 and <0.2 mg L^{-1}. In terms of pH, Al release was found to be most significant at pH 3 with values of ~7 mg L^{-1} [20] and >20 mg L^{-1} [32] being reported. Furthermore, the review shows that the concentration of leached Al was higher at the beginning of the experiments in some cases, but this became lower in the course of the experiment.

[Insert Table 1 here]

The impact of Al release was evaluated in some cases with mixed conclusions obtained and varied depending on the focus on the impact. For instance, Mortula and Gagnon [31] reported Al leaching in their study but concluded that the level of Al in the leachate was within reasonable range for surface water disposal and the level is not high enough to cause toxicity for aquatic species if disposed. Agyin-Birikorang et al.[10] also concluded that Al-WTS can be safely used to reduce P leaching into groundwater without increasing Al concentration of the groundwater. However, although Kaggwa et al.[4] in a study on the discharge of Al-WTS to a natural wetland reported that there was no adverse effect on the water quality and the growth and development of the aerial biomass, Al toxicity in the rooting system was reported to cause root abnormalities.

[INSERT FIG. 1 HERE]

3. Investigation of Al release from a pilot field-scale Al-WTS based CWs

3.1. CWs description and monitoring of Al release

The development of the novel CWs employing Al-WTS as main substrate for wastewater treatment has been extensively conducted in the Centre for Water Resource Research, University College Dublin, Ireland. Zhao et al. [42] gives a roadmap of the different phases leading to the development of the system. Our previous investigation has shown that the dominant component of the Al-WTS is aluminum with ~46% in mass expressed as Al_2O_3. The other principal chemical components also include Fe^{3+}, Ca^{2+}, Mg^{2+}, Cl and SiO_4 while morphological analysis shows that the Al-WTS is amorphous.

After extensive laboratory scale studies, a pilot field-scale CWs employing Al-WTS as main substrate was set up to treat wastewater emanating from an animal farm. The CWs consists of four stages and each stage was constructed using similar 1100L plastic bins with dimensions of 108cm $\times$ 95cm $\times$ 105cm (L $\times$ W $\times$ H). The Al-WTS cakes used in the CWs were collected from the filter press of the sludge dewatering unit of a Water Treatment Works in Southwest Dublin, Ireland where aluminium sulphate is used as coagulant. Detailed description of the system and analysis of its performance so far has been reported in Zhao et al.[42]

During the first year of operation of the system, a designed hydraulic loading rate of 0.29 $m^3m^{-2}.d^{-1}$ (where m^2 represents the total surface area of the CWs) was applied. Samples of influent and effluent were collected weekly for the first 4 weeks and every 2 weeks thereafter (except for the period between the 12th and 18th week when the system was stopped to allow for some changes on the farm) and analysed for total- and soluble Al. Al analysis was carried out using a Hach DR/2400 spectrophotometer according to its standard operating procedures. Soluble (dissolved) Al was operationally defined as samples filtered through 0.45μm Millipore membrane filter. Other water quality parameters including COD, BOD_5, TN, NH_3-N, NO_3-N, NO_2-N, P, SS and Turbidity were also analysed to assess the treatment performance and efficiency of the CWs.

3.2. Results

Detailed results of the field performance of the CWs are outside the scope of this paper. However, in brief, the system showed great promise as a low-cost system of choice for effective removal of pollutants from wastewater. During the first year of operation, the mean monthly removal efficiencies obtained was determined to range from 56.6%-83.5%, 35.6%-84.2%, 11.2%-77.5%, 48.5%-92.5%, 75.4%-93.8%, 73.0%-96.5% and 46.3%-83.3% for BOD_5, COD, TN, NH_3, TP, P and SS. Furthermore, the system showed distinct P removal which was high and sustained from the beginning of the trials and also, the system was effective in reducing levels of ammonia-nitrogen in the influent.

Levels of total and soluble Al monitored in the influent and effluents of the individual stages of the CWs during the first year of operation are shown in Fig. 2. The levels of total and soluble Al were mostly higher in the effluents than in the influent and this indicates release of Al from the Al-WTS into the treated effluent. However, it can be observed from the figure that the highest level of total and soluble Al were detected during the first three weeks of operating the system and decreased afterwards except for the 26th week when a rise in the level of both parameters was observed. Beyond the first three weeks, concentrations of total and soluble Al in the effluent remained below the recommended guideline limit of 0.2 mg L^{-1} for drinking water standard and effluent discharge.[43-46]

Al was also detected in the influent wastewater into the CWs. By accounting for this background Al concentration in the wastewater being treated, the level of Al leached in each stage of the CWs was determined and presented in Fig. 3. The levels of Al leached were generally low and range from 0.02 to 0.06 mg L^{-1} across the stages. It is also noted that leached levels of Al increased across the stages from the 1st stage to the 4th stage.

[INSERT FIG. 2 HERE]

[INSERT FIG. 3 HERE]

Fig. 4 shows the pattern of P removal and Al release in the individual stages of the CWs. An inverse relationship was observed but regression analyses indicate a weak linear relationship with R^2 << 0.5. From Fig. 4, it can be seen that whereas P concentration decreased from the influent and across the stages, both total and soluble Al showed an opposite trend with total and soluble Al concentrations increasing from the influent and across the stages. It can also be deduced from the figure that while the 1st stage accounted for most of the P removal, it had the least concentration of total and soluble Al in its effluent compared to the other stages. On the other hand, the last stage (i.e. 4th stage) accounted for the least P removal but it had the highest concentration of total and soluble Al in its effluent relative to the other stages. It therefore follows that as more P is being removed mainly through adsorption on the Al-WTS, the tendency for leaching out of Al is reduced.

[INSERT FIG. 4 HERE]

4. Discussion

The reuse of Al-WTS as a substrate in CWs represents an innovative approach to enhancing wastewater treatment. However, due to the significant amount of Al contained in the Al-WTS, there is appreciable concern as to Al release during such reuse and its environmental impact. The pilot field-scale CWs demonstrated in this study represents the first of its kind employing Al-WTS as the main substrate. Therefore, there is no reported study which has evaluated Al release in such field scale CWs. The desk review conducted in this study indicates that while Al leaching is reported in some studies (mainly laboratory based studies), a major conclusion is that the Al concentration in the leachate does not pose significant environmental risk. The desk review further reveals that Al leaching in most of the studies is mainly pH dependent and Al in Al-WTS is in a stable form which may not be readily available and leached.[40]

Several regulations have been promulgated in relation to Al concentration in drinking waters and effluents for discharge. In Ireland and UK, the prescribed limit for Al discharge into all waters is 0.2mg L^{-1}.[43-46] For drinking waters, the World Health Organization suggests a maximum limit of 0.2 mg L^{-1} [44] while in the USA, the United States Environmental Protection Agency secondary drinking water regulation stipulates a range of 0.05–0.2 mg L^{-1}.[45] The results obtained in this study shows that the level of Al leached were quite low ranging from 0.02 to 0.06 mg L^{-1} and the overall concentration of Al in the effluents were well below the general regulatory guideline limit of 0.2 mg L^{-1} except for the first three weeks of operation. It is noted that the wastewater being treated had some background Al concentration and this may have contributed to the overall concentration of Al in the effluents. It is therefore very important to consider the background Al concentration of the wastewater being treated so as not have an exaggerated view of Al release from the Al-WTS during such reuse. However, even though the levels of Al found in the effluents do not represent an imminent environmental or health risk, periodic determinations are advisable.

In a previous study of four laboratory scale CWs employing Al-WTS as substrate in different proportions, it was found that the concentration of total and soluble Al in the effluent were above the prescribed limits for discharge in most cases, especially at the beginning of the experiments.[12] In particular, levels of soluble Al monitored in the effluents were reported to range from 0.058 mg L^{-1} to 1.106 mg L^{-1}. However, the wastewater being treated had some background Al concentration which would have contributed to levels of Al obtained. Interestingly, further analysis of the leachates from the study indicated that Al exhibited the least leaching potential relative to the initial content in the fresh Al-WTS.

The initial high concentration of Al in the leachate during the first three weeks may have been influenced by hydrolysis. Yang et al.[40] did a study on the mechanism and characteristics of P adsorption onto Al-WTS by focusing on the pre-hydrolysis process and the adsorption process. It was shown that Al-WTS exhibited a strong hydrolysis potential which was characterized by an initial rapid release of several ions and total Al in the first 24 h followed by a slow release. Total Al was reportedly increased from 0 to 0.033 mg L^{-1} during the hydrolysis but this was found to decrease to 0.023 mg L^{-1} during the P adsorption stage. Thus it could be seen that there is a potential for some Al release during the hydrolysis process of the Al-WTS and this may have influenced the initial high concentration of Al found in the effluent during the first three weeks of operation.

On the effect of P adsorption on Al release from the Al-WTS, result obtained in this study indicates that P adsorption onto the Al-WTS may have served to reduce Al leaching from the Al-WTS. Previous researchers have shown that P adsorption is through a kind of inner-sphere complex reaction. It is hypothesized that P adsorption occurs at the WTS–hydrous aluminium oxide interface with phosphate replacing singly coordinated hydroxyl groups and then reorganizing into a very stable binuclear bridge between cations.[22, 40] Analysis of P removed and Al released in each stage of the CWs reveals an inverse trend between P adsorbed and Al released across the four stages of the CWs. The lower the P concentration in the effluent, the higher the Al concentration but regression analysis shows that the relationship is not strongly correlated. It therefore follows that P adsorption onto the Al-WTS may contribute to reducing Al leaching from the Al-WTS, but it is not a direct relationship. Mortula and Gagnon [31] reported similar findings in their studies and also suggested that P adsorption on Al-WTS may have an insignificant effect on Al release.

5. Conclusions

Environmental and health concerns about the possible release of Al during the operation of a novel CWs configured using Al-WTS as the main substrate were addressed in this study. Levels of total and soluble Al were particularly monitored over 42 weeks during the operation of the pilot field-scale CWs. Results indicate that although Al release was observed, the level of Al released in the effluent was quite low and ranged between 0.02 to 0.06 mg L^{-1}. The concentration of total and soluble Al monitored in the effluents were all below the general regulatory guideline limit of 0.2 mg L^{-1} for drinking water and effluent discharges, except during the first 3 weeks of operation. An inverse trend was observed between P reduction and Al release across the stages of the CWs, indicating that P adsorption onto the Al-WTS may serve to reduce Al release. However, regression analysis indicates a weak relationship. Overall, the Al-WTS based CWs showed great promise for pollutants removal (particularly P). Although the release of Al was observed, it does not pose any imminent environmental and health risk. However, periodical monitoring is recommended.

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Figure captions:

Fig. 1 Reported levels of Al released/leached during studies on the reuse of Al-WTS

Fig. 2 Monitoring results for Al concentration in the influent and effluent of the CWs showing (a) total Al and (b) soluble Al

Fig. 3 Mean levels of influent Al and leached Al in the different stages of the CWs

Fig. 4 Trends of P reduction and Al release from the influent and across the stages of the CWs (mean values plotted)

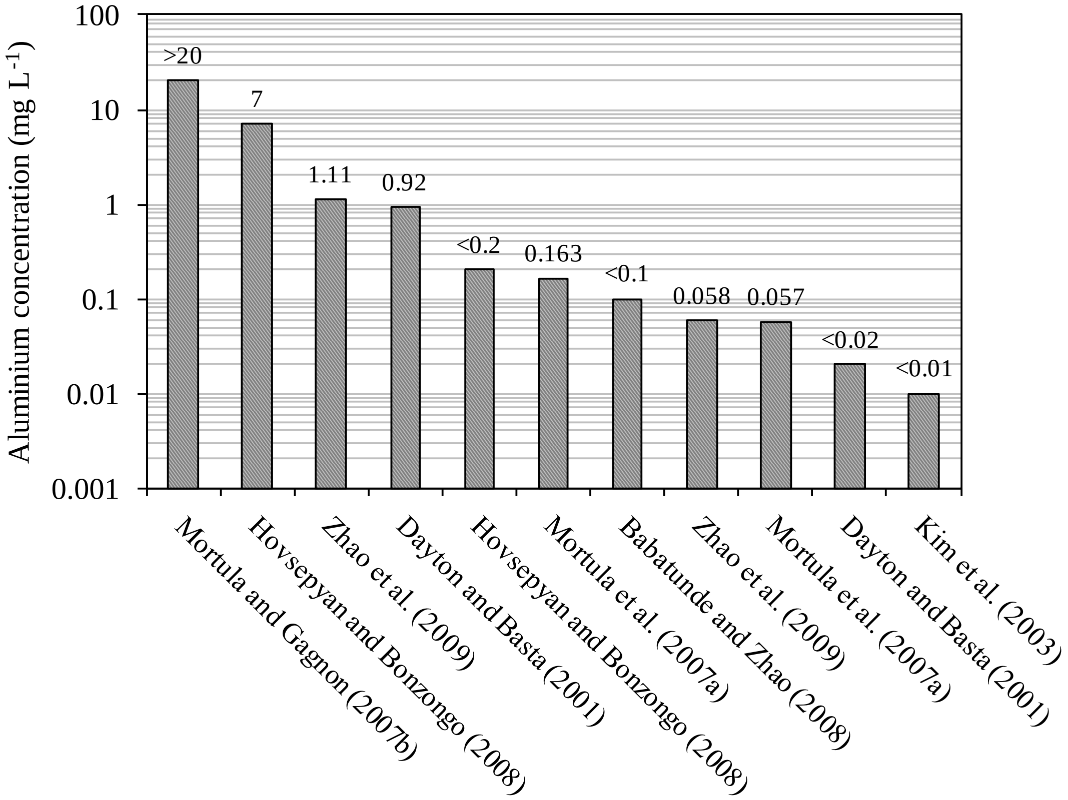

Aluminium concentration (mg L-1)
100
10
1
0.1
0.01
0.001
>20
7
1.11
0.92
<0.2
0.163
<0.1
0.058
0.057
<0.02
<0.01
Mortula and Gagnon (2007b)
Hovsepyan and Bonzongo (2008)
Zhao et al. (2009)
Dayton and Basta (2001)
Hovsepyan and Bonzongo (2008)
Mortula et al. (2007a)
Babatunde and Zhao (2008)
Zhao et al. (2009)
Mortula et al. (2007a)
Dayton and Basta (2001)
Kim et al. (2003)

Fig. 1

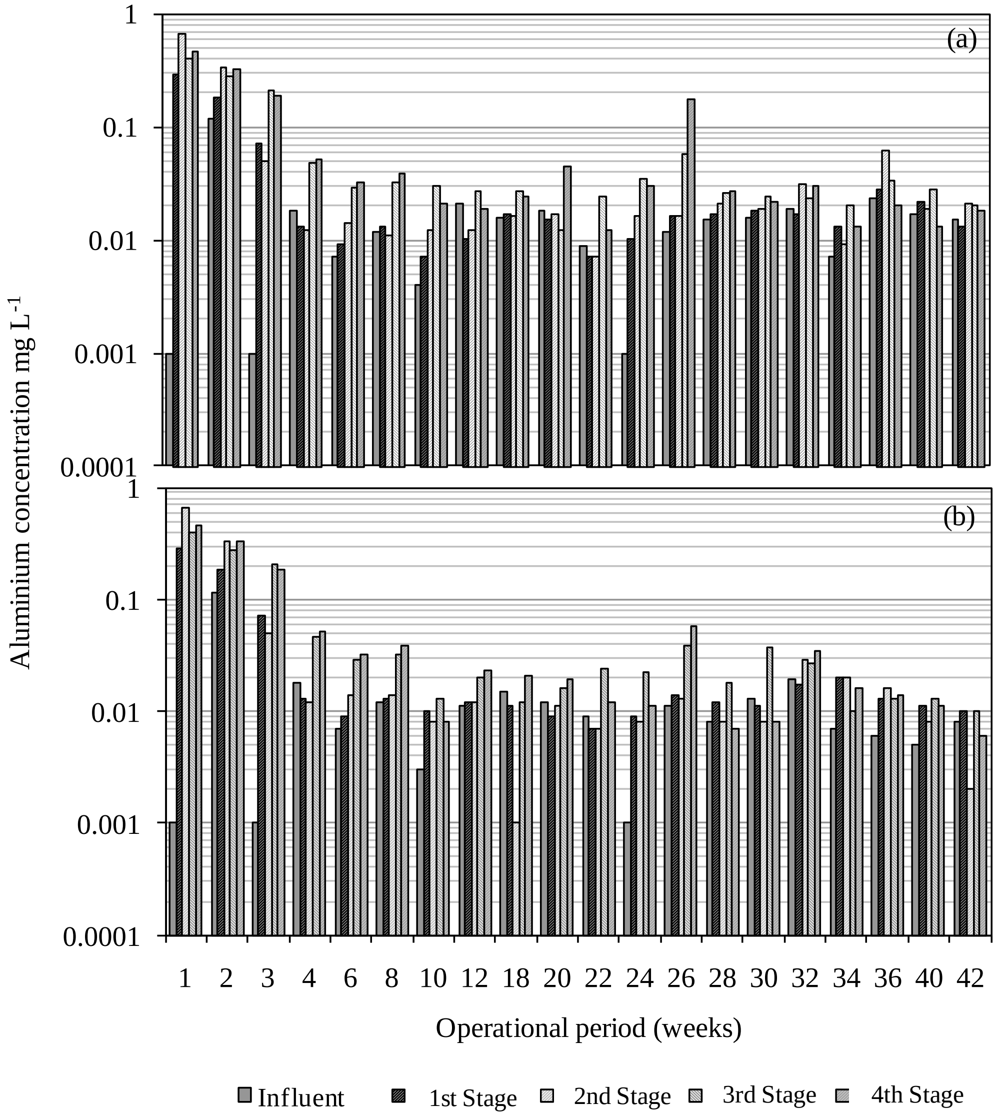
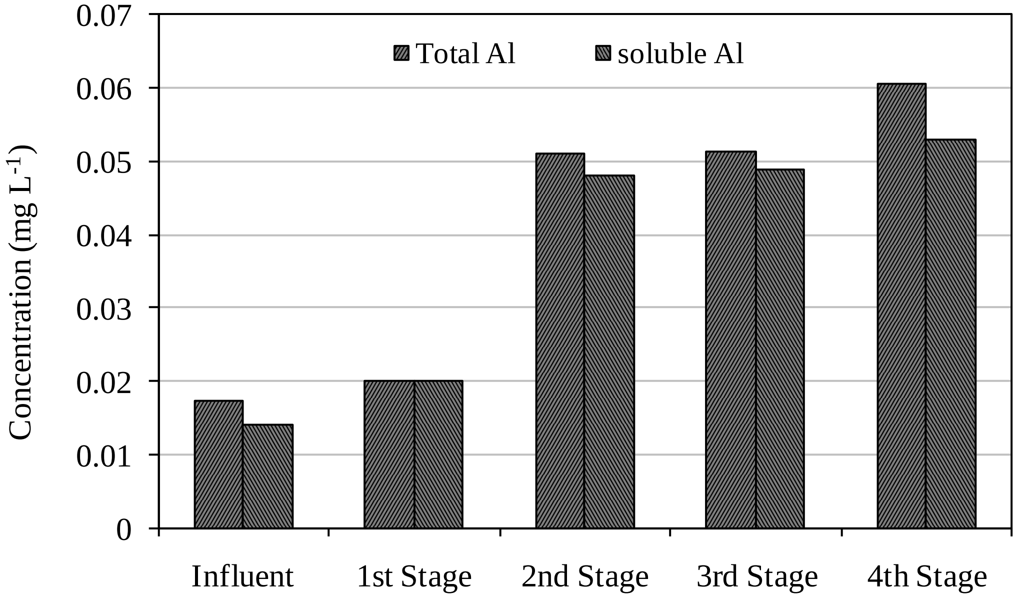

(a)
(b)
1
0.1
0.01
0.001
0.0001
Aluminium concentration mg L-1
1 2 3 4 6 8 10 12 18 20 22 24 26 28 30 32 34 36 40 42
Operational period (weeks)
Influent
1st Stage
2nd Stage
3rd Stage
4th Stage

Fig. 2



0.07
0.06
0.05
0.04
0.03
0.02
0.01
0
Total Al
soluble Al
Concentration (mg L-1)
Influent
1st Stage
2nd Stage
3rd Stage
4th Stage

Fig. 3

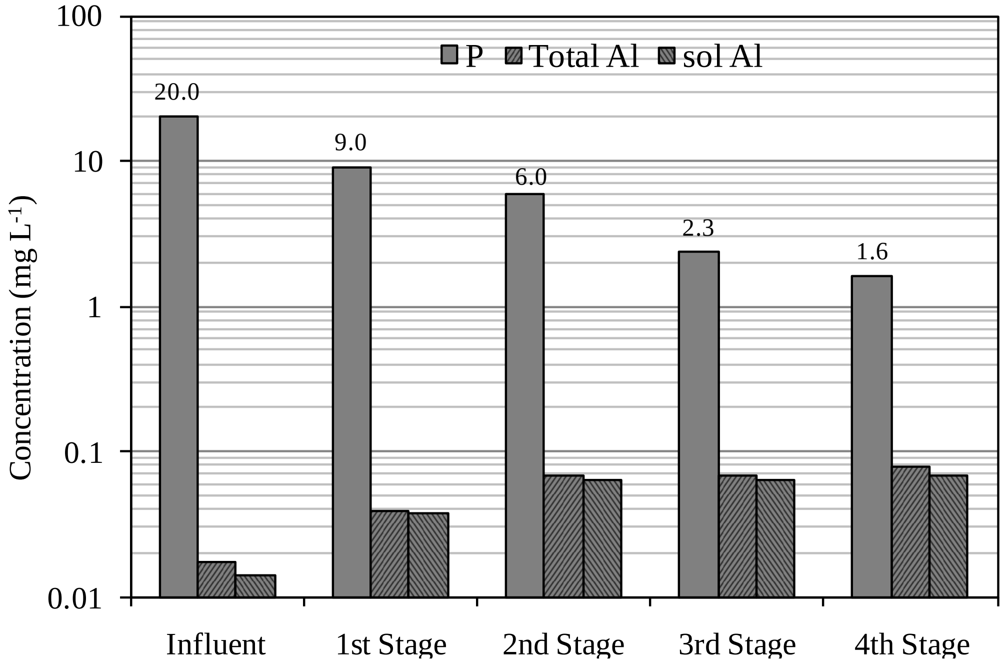

100
10
1
0.1
0.01
Concentration (mg L-1)
P
Total Al
sol Al
20.0
9.0
6.0
2.3
1.6
Influent
1st Stage
2nd Stage
3rd Stage
4th Stage

Fig. 4

Table 1. A review of findings related to leaching or release of Al from studies focusing on reuse of Al-WTS

Study type	Study objectives	Findings related to Al leaching/release	References
Field study	Direct evaluation of the impacts of land application of Al-WTR on ground water quality.	It was concluded that for the study period, Al-WTS can be safely used to reduce P leaching into ground water without increasing the Al concentration of ground water.	10
Laboratory scale	An investigation into extending the reuse of Al-WTS as an adsorbent for condensed P.	It was reported that loss of solids was minimal and the Al content in the treated effluent remained below 0.1 mg-Al^{3+} L^{-1}.	11
Laboratory scale	The studies were aimed at extensive characterization of Al-WTS as a potential substrate in engineered wetlands and its trial as a media in four different configuration of laboratory scale wetlands	The Al-WTS was characterized as being mainly composed of amorphous aluminium.	2
Laboratory scale	To investigate Al leaching from laboratory scale constructed wetlands using Al-WTS as substrate	Effluent dissolved levels of Al ranged from 58 μg L^{-1} to 1,106 μg L^{-1}	12
Laboratory scale	To evaluate the technical and economical viability of using WTSs as a recycled material in concrete, cement mortars, clay materials and geotechnical works	Leaching solution of the WTS was found to be alkaline and poorly mineralised and the incorporation of the WTS did not promote any contamination. There was however no particular mention or focus on Al leaching/release.	13
Laboratory scale	To recycle Al-WTS using a chemical precipitation process to promote the removal of lead metal in wastewater	Increased desorbed Al concentration was reported	14
Laboratory scale	To characterize WTSs and determine their potential for use as soil substitutes	Soluble aluminium levels in the WTSs were found to range from 0.02 to 0.92 mg L^{-1}. No conclusion on leaching was given	15
Laboratory scale	The study objective was to determine WTS component responsible for P sorption and reduction of P in runoff water using 21 Al-WTSs.	Al leaching/release not particularly mentioned nor studied.	16
Batch study	Freshly precipitated spent Al-WTS and alum were tested and compared for their efficiency to remove phosphorus in synthetic wastewater	Al leaching/release not particularly mentioned nor studied	17
Laboratory scale	The study examined four WTSs from North America to determine the coagulant role in phosphate adsorption by the WTSs	Al leaching/release not particularly mentioned nor studied.	18

Table 1. Cont'd

Study type	Study objectives	Findings related to Al leaching/release	References
Laboratory scale	To study the feasibility of reusing Al-WTS to improve particulate pollutant removal from sewage	Al leaching/release not particularly mentioned/studied. However, reference to earlier works by the same authors shows that few soluble aluminium ions were found in the Al-WTS reuse tests.	19
Laboratory scale	To assess the potential of WTSs to sorb and immobilize Hg from aqueous solution in a series of batch experiments	The dissolution of Al was found to be significant at pH 3 (~ 7 mg L^{-1}), and at pH ≥ 4 concentration of Al were either lower than the 0.2 mg L^{-1} limit or below the detection limit (3μg L^{-1}). However, the leachable Al concentration obtained using the synthetic precipitation leaching procedure test was below the local groundwater guideline limit of 0.2mg L^{-1}.	20
Laboratory scale	Study was conducted to determine factors affecting the leaching out of metals (Al, Fe, Ca, Mg, Mn) from WTS	It was reported that Al, Fe and Ca are generally immobile in WTS but these metals will leach out quickly when the pH < 4.	21
Laboratory scale	To study P adsorbing mechanism(s) of Al-WTS	Increased desorbed Al concentration was reported	22
Laboratory and field studies	The study examined the effect of Al-WTS discharge on a natural wetland	Al-WTS was found to have no adverse effect on water quality of the swamp and also growth and development of aerial biomass in *Phragmites mauritianus* despite the resultant lowering of phosphorus concentrations in the Phragmites stand. Sediment chemistry was reportedly affected by the discharges but not to any considerable effect. However, Al toxicity in the rooting system was reported to cause root abnormalities.	4
Laboratory scale	To test alum sludge as an inexpensive alternate adsorbent for various P species in wastewater.	Al release was observed. Al concentration of the solution had lowest values around pH 6 for inorganic phosphates. At pH > 6, greater amount of Al was dissolved by the adsorption of inorganic phosphate than in the controlled set. In column experiment, Al concentration was found to be below 0.01mg L^{-1} for pH 4 and 5 while for pH 3 and 12, it was noted that the Al concentrations could be a concern.	23

Table 1. Cont'd

Study type	Study objectives	Findings related to Al leaching/release	References
	To evaluate the P sorption and desorption potential, and the physicochemical characteristics of several materials including the WTSs as a management practice option to reduce P loss from soil to water	Al leaching/release not particularly mentioned nor studied.	24
Laboratory scale	A greenhouse experiment was conducted to quantify the effects of Al-WTS on bioaccumulation of some heavy metals in plant tissue and to determine the effects of the Al-WTS on soil Al and Al phytotoxicity for the corn plants in alkaline soils.	Application of Al-WTS was reported not to cause aluminum phytotoxicity symptoms.	25
Laboratory scale	The study evaluated the ability of WTSs to adsorb As(V) and As (III)	Al leaching/release not particularly mentioned nor studied.	26, 27
Laboratory scale	Al-WTS was incorporated into the manufacture of red ceramics and the influence of firing temperature on the technological properties of the red brick was evaluated	Al leaching/release not particularly mentioned nor studied.	28
Laboratory scale	To determine the leachability of aluminium from residuals generated from phosphorus treatment using different types of wastewater on Al-WTS. Leachate concentrations as obtained from Toxicity characteristics leaching procedure (TCLP) tests were examined to evaluate the leaching potentials in land based disposal options.	(1) Low amounts of Al (0.057-0.163 mg L^{-1}) were leached from the residuals mixed with surface water. (2) Phosphorus adsorption can make physical and chemical changes to Al-WTS and these can affect the leachability of Al-WTS. (3) TCLP tests result indicates that phosphorus treatment of Al-WTS showed a reduction in aluminium leachability ranging from 54% to 97%. (4) Dried Al-WTS has a lower tendency to release aluminium and manganese than raw Al-WTS	29
Batch and fixed bed column tests	The study was aimed at investigating the effectiveness of Al-WTS (oven dried) for the adsorption of phosphorus from aquaculture process water. Batch adsorption tests and fixed bed column tests were used	Al leaching was observed and leachate value was reported to be < 0.5mg L^{-1}. Al leaching was consistently high at the beginning of experiments but became lower in the course of experiments. Highest and lowest leaching were reported to occur at pH's 3 and 5, respectively. It was concluded that Al level in the leachate was within reasonable range for surface water disposal and the level is not high enough to cause toxicity for aquatic species if disposed.	30, 31

Table 1. Cont'd

Study type	Study objectives	Findings related to Al leaching/release	References
Batch and fixed bed column tests	The study was aimed at investigating the effectiveness of Al-WTS (oven dried) for the adsorption of phosphorus from secondary municipal effluent. Batch adsorption tests and fixed bed column tests were used	Al leaching was observed and it was reported to be high (> 1 mg L^{-1}) at the beginning of the experiments for both pH's 5 and 7, and decreased to < 0.2 mg L^{-1} in the course of the experiment. It was concluded that Al level in the leachate was not high enough to cause toxicity for aquatic species if disposed.	32
Laboratory scale	A laboratory experiment was performed to assess the leaching of dried Al-WTS to five lake water samples. Tests were also done to evaluate the effect of pH levels (4, 5.5 and 7) and drying mechanism of Al-WTS (oven, air or freeze-thaw).	It was inferred from the results obtained that low background Al concentrations has an effect on the increasing leachability of Al from oven-dried Al-WTS. In one of the samples studied, it was further observed that the addition of oven-dried Al-WTS decreased the Al level in the Lake sample with an exception at pH 4. Furthermore, it was reported that high Al concentrations in uninterrupted lake water were eventually adsorbed onto Al-WTS. Overall, it was reported that changes in pH did not affect the leachability of oven-dried Al-WTS and drying of Al-WTS did not affect the Al leachability.	7
Batch and fixed bed column tests	Fixed bed columns were used to evaluate the effectiveness of P adsorption on oven dried alum residual solids with emphasis on the effect of key operating parameters (pH, particle size, and initial P concentration) on effectiveness of P removal.	Al leaching was found to be higher than 20mg L^{-1} for an influent pH level of 3.	33
Review	A review was undertaken to assess the feasibility of various disposal options for WTSs	A major finding was that the presence of active aluminium or iron hydroxides is often quoted as being a potential problem when WTSs is reused in agriculture and horticulture, but the author argued that this should not restrict the reuse of the WTS since many tropical soils have a high percentage of free iron and aluminium hydroxides	34
Laboratory scale	To evaluate the feasibility of employing some locally available oyster shells (OS) Al-WTS as the P adsorption media of constructed wetland beds	Al leaching/release not particularly mentioned nor studied	35

Table 1. Cont'd

Study type	Study objectives	Findings related to Al leaching/release	References
Laboratory scale	The study aimed to investigate the technical feasibility of integrated constructed wetland system consisting of a pre-filter unit and a constructed wetland in series, packed with Al-WTS and oyster shells as the filter media respectively, for nitrogen and P removal from domestic wastewater.	Al leaching/release not particularly mentioned nor studied.	36
Laboratory scale	The study aimed to evaluate the effect of WTS type and application rate on As immobilization in two soils with contrasting physicochemical properties and to determine the As desorption potential in the presence of WTSs from the As loaded soils using high rates of common fertilizer P	Al leaching/release not particularly mentioned nor studied.	37
Laboratory scale	To examine the reuse of Al-WTS as an adsorbent for the removal of fluoride from polluted waters using raw and treated Al-WTS in flask shaking experiments	Not mentioned	38
Laboratory scale	To carry out physiochemical characterization of the WTSs and assess their potential for land application.	Exchangeable Al ranged from 0.05 to 0.07 cmol kg^{-1}. It was suggested that the low exchangeable aluminium values which reflect very low acid producing potential is as a result of the lime component of the WTSs and their neutral to alkaline pHs. It was suggested that the WTSs have potential for land application but an exception may be where extreme acidity may render their neutralising capacity ineffective.	39
Laboratory scale	An investigation into the mechanism and characteristics of P adsorption onto Al-WTS	A major conclusion from the study is that Al in the alum sludge is in a stable and immobilized form at the test pH range of 5.98-7.21.	40
Laboratory scale	To explore novel application of dewatered alum sludge as a main substrate in a single model reed bed to treat P rich animal wastewater on a short term basis.	The possibility of substance release during such reuse was highlighted	41