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Distribution of Heavy Metals in Vegetative Biofiltration Columns

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Abstract: This study investigated the distribution of heavy metals in vegetative biofiltration columns irrigated by synthetic greywater. Twelve species of ornamental plants (three plants from each species) were planted in the same designed 36 biofiltration columns. Samples of effluent water, soils, roots, shoots and leaves were collected and analyzed. It was observed that before irrigation, the distribution of copper was in soils (0%), roots (42%), leaves (37%) and shoots (21%). After irrigation, this distribution changed to soils (29%), roots (39%), leaves (17%) and shoots (15%). It was found that lead concentrations decreased in soils from (84% to 7%), but increased in plants (from 16% to 93%) following irrigation with greywater. In contrast, the distribution of zinc changed from leaves (46%), roots (22%) and soils (16%) before irrigation to 89% in leaves and soils and 11% in shoots following irrigation. The chromium distribution before and after irrigation was found to be almost unchanged in soils, shoots and effluent water, but it increased in roots (19.4% to 26.9%) and decreased in leaves (11.4% to 5.8%). The outcomes of this study demonstrated that heavy metals mostly accumulate in soils and roots, and it is necessary to investigate their potential detrimental effects on the receiving environment.

Keywords: biofiltration; vegetation; heavy metals; greywater

1. Introduction

Biofiltration systems are shallow landscaped depression areas, and their pollutants removal mechanisms from influent stormwater or wastewater rely on engineered soils, enhanced vegetation and biofiltration processes [1]. Biofiltration systems provide an aesthetic environment to urban landscape. The upper soil layer of biofiltration system, which is popularly known as the mulch layer, promotes microbial growth for biodegradation of hydrocarbons and organic matter, and helps with filtration process [1]. The mulch layer is followed by an underlying soil layer, which helps infiltration and serves as a growing medium for plants. The clay contents of soil layer provide an adsorption medium for pollutants such as heavy metals, hydrocarbons and nutrients [2]. An underlying sand bed layer, which is the largest component, follows the planting soil layer and facilitates drainage of influent

water. This layer also provides a polishing treatment through aerobic reactions. The effluent drained out of the system is collected through a drainage facility, and can end up with either groundwater recharge or discharge as stream baseflow. The vegetation in biofiltration systems offers two vital biological processes, namely, a phytoremediation process by which plants uptake pollutants and a bioremediation process involving microbial conversion of nitrogen [3]. Roots provide several beneficial functions to the biofiltration system such as their support of many microorganisms such as bacteria and fungi, improvement of hydraulic conductivity of soil, effect upon the rhizosphere characteristics and enhancement of plants' nutrient intake and respiration capacity [4].

Investigation of heavy metals in biofiltration systems is an important issue because of their possible accumulation in the system and their long-term effects on the environment [5]. According to Geronimo et al. [6], the removal mechanisms for heavy metals in biofiltration systems are less complicated compared to those for nitrogen and phosphorus compounds. Biofiltration systems have been found to reduce the influent lead (Pb) concentration by between 83% and 90%, whereas reductions of 62% to more than 90% have been observed for zinc (Zn) [7–9]. Copper (Cu) removal efficiency has been more variable. Although Davis [8] reported a 57% reduction of Cu, Hatt et al. [7] observed a reduction of more than 90%. On the other hand, Li and Davis [10] and Trowsdale and Simcock [9] noticed an increased concentration of Cu in biofiltration effluents. Unlike Zn and Pb, Cu associates with organic matter in the soil and is likely to dissolve in the organic-rich water that is commonly found in biofiltration systems [10,11]. Additionally, Trowsdale and Simcock [9] observed an increased dissolved Cu concentration with an increase in outflow volume, signifying that the flushing of dissolved Cu is related to the flow rate. Pb exhibits a very low mobility (ability to penetrate down through soil media) compared to the moderate mobility of Cu and Ni. However, mobility of cadmium (Cd) is much higher compared to Cu and Ni. Consequently, Cd poses a higher risk of being discharged from the system [12].

Heavy metals in biofiltration systems are usually adsorbed within the upper soil layer (10 to 15 cm) and their concentration decreases with an increase of depth [10,13]. Soil generally possesses a limited sorption capacity, thereby making it possible for heavy metals to accumulate to a level detrimental to the ecosystem, usually after around 15 to 20 years of operation [5]. Sun and Davis [14] observed that bio-accumulation of metals in biofiltration systems is higher in the roots compared to the shoots of the grasses they studied. Their research also showed that soil media captures between 88% and 97% of the metal inputs, whereas only 0.5% accumulates in the plants, and the remaining 2% to 11.6% escapes out of the system. Several previous studies have confirmed that different plants exhibit a different capacity to accumulate heavy metals in biofiltration systems [15–18].

Recycling and reuse of greywater through a vegetative biofiltration system is a comparatively new idea in urban water management. Recent studies have shown that vegetated systems (biofiltration, living wall systems, for example) are effective in reducing greywater pollutants [19–21], but none of these studies have investigated the removal and distribution of heavy metals in the systems. The presence of heavy metals in greywater and their potential effects when reused for irrigation purposes have been reported in several previous studies [22–27]. Long-term greywater-based irrigation could lead to the accumulation of metals in soils and may exceed acceptable environmental levels [24,26]. The metals not only accumulate in the irrigated soil, but can also accumulate in plants [24] and could end up increasing the metal concentrations in groundwater [26]. How much of the metals are adsorbed by soils and the amount of uptake by plants are important research questions. In terms of plant uptake, it is important to understand how much of the metals are stored in roots, shoots and leaves.

This study investigated the distribution of heavy metals in the different components of biofiltration systems. Specific objectives were to observe the distribution of heavy metals in soils, plants (roots, shoots and leaves) and in effluent waters before and after irrigation with greywater. Chowdhury et al. [20] reported the concentrations of heavy metals in effluent waters; therefore, this study is an advancement in its considering of metals concentrations in soils and plants, as well as its provision of a complete mass balance of heavy metals in vegetative biofiltration columns. Green infrastructures such

as the vegetative systems are becoming very popular and essential elements of the urban landscape design of modern cities. Therefore, outcomes of the study may be useful for the selection of appropriate plants for vegetative systems and to explore the fate of metals from the systems.

2. Materials and Methods

The study was conducted in the arid climate of the city of Al Ain, United Arab Emirates (UAE). Thirty-six laboratory scale biofiltration columns were constructed. Description of columns used in this study and the experimental set-up scheme are available in Chowdhury et al. [20]. The columns were built using polyvinyl chloride (PVC) pipe, and a faucet was connected at the bottom of the columns for effluent sample collection. The length and diameter of the column were 60 cm and 15.8 cm, respectively. The effective length of the columns was 54 cm, with a 6 cm depth being left for irrigation water ponding. There were three layers in the columns, a 40 cm deep filter media (root growth layer) at the top, followed by a 8 cm deep transition layer and a 5.3 cm deep drainage layer at the bottom. The filter media contained a well-graded soil of particle size 0.075 to 4.75 mm (25% sand, 20% black soil, 35% gravel of particle size 0.5 to 2 mm and 20% gravel of particle size 2 to 5 mm). The transition layer contained soil of particle size 0.5 to 2 mm, whereas drainage layer contained well-graded aggregates of size 2 to 5 mm.

Twelve ornamental plant species, which are widely used for landscaping and gardening in the study region (UAE), were selected for this study. A total of 36 plants, three plants from each species (3 plants × 12 species = 36), were planted in the 36 columns. The selected 12 plants were *Alternanthera ficondea* (Alt), *Canna indica* (Can), *Dodonaea viscose* (Dod), *Ficus nitida* (Fic), *Hibiscus rosa-sinensis* (Hib), *Ixora coccinaea* (Ixo), *Jasmine sambac* (Jas), *Lantana camara* (Lan), *Pennisetum seateceum* (Pen), *Rhoeo discolor* (Rho), *Vinca rosea* (Vin) and *Vitex agnus* (Vit).

The plants in the columns were nursed for 2 months (September and October 2015) using municipal water irrigation. Equal amount of synthetic greywater (6.4 mm to 25.5 mm per day) was applied uniformly to all columns whereas the bottom faucets were kept open for allowing free drainage of water without any detention. The study was conducted from September 2015 to May 2016. The root and soil samples were collected twice, once at the beginning (1 September 2015) and once at the end of the experimental period (5 May 2016). Effluent water samples were collected fortnightly from 5 November 2015 to 5 May 2016 (13 samples), whereas plant leaf and shoot samples were collected monthly, and thus there were seven experiments on leaves and shoots.

The composition of the synthetic greywater conformed to Diaper et al. [28]. Heavy metals in synthetic greywater were kept within the ranges reported in previous studies [22,23,29]. All the samples were prepared and analyzed using inductively coupled plasma-optical emission spectrometry (ICP-OES), SW-846, method 6010D [30]. Five heavy metals were analyzed in this study, these being cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb) and zinc (Zn).

The analysis of variance (ANOVA) available in Microsoft Excel 2013 was applied for the analyses of concentration of heavy metals, as described in Chowdhury et al. [20]. For each heavy metal, plant species were represented by row (horizontal axis) and columns (vertical axis) in the ANOVA matrix representing their three replicates. A 5% significance level was considered (null hypothesis = no significant difference in mean values).

3. Results and Discussion

3.1. Heavy Metals in Effluent Water

Figure 1 shows the average concentrations of metals in effluent water samples before and after irrigation with greywater. It was observed that Pb is the predominant metal in the effluent water samples, followed by Cu and Cr. Except for columns with the plant *Dodonaea viscose*, an increased Cd concentration was observed in effluent water samples from all columns at the end of the experiment. Comparatively lower concentrations of Cd and Zn were present in the effluent water samples, both at

the beginning and at the end of experiment. Pb is adsorbed rapidly to form a stable and sparingly soluble compound [11]. As a result, $Pb(OH)_2$ and $Pb(OH)^+$ compounds formed from hydrolysis reactions could be drained out with effluent waters [31]. In addition, the reaction of Pb with solid surfaces occurs, but becomes slower with time [11]. This, together with the continuous addition of Pb in the system through greywater irrigation, could explain the observed increases of Pb concentration in the effluent water samples at the end of the experiment. Chowdhury et al. [20] provided the detailed analysis of heavy metals in effluent water samples.
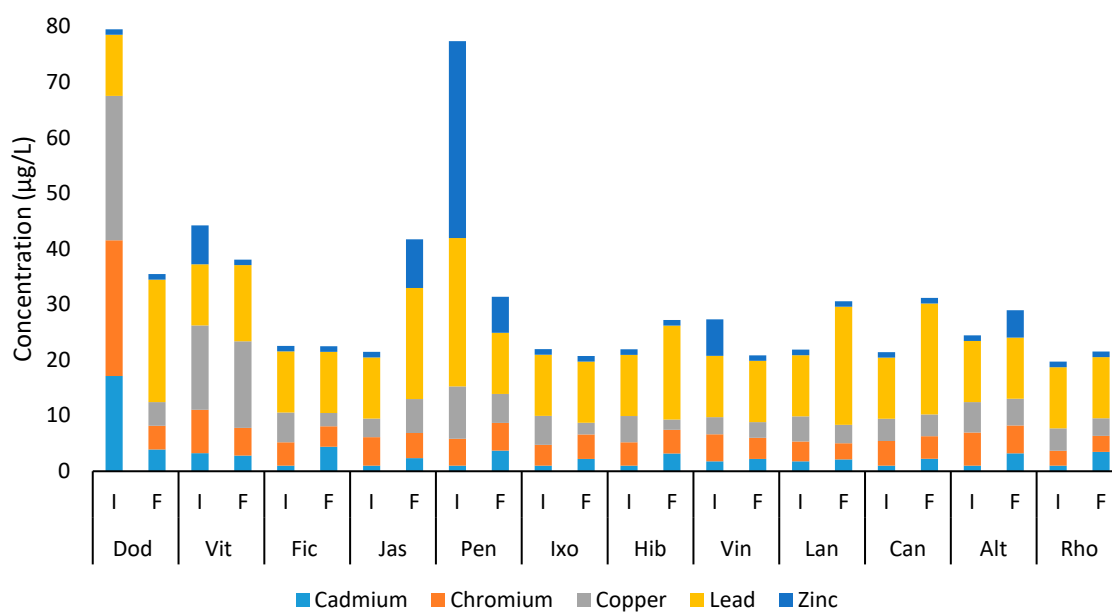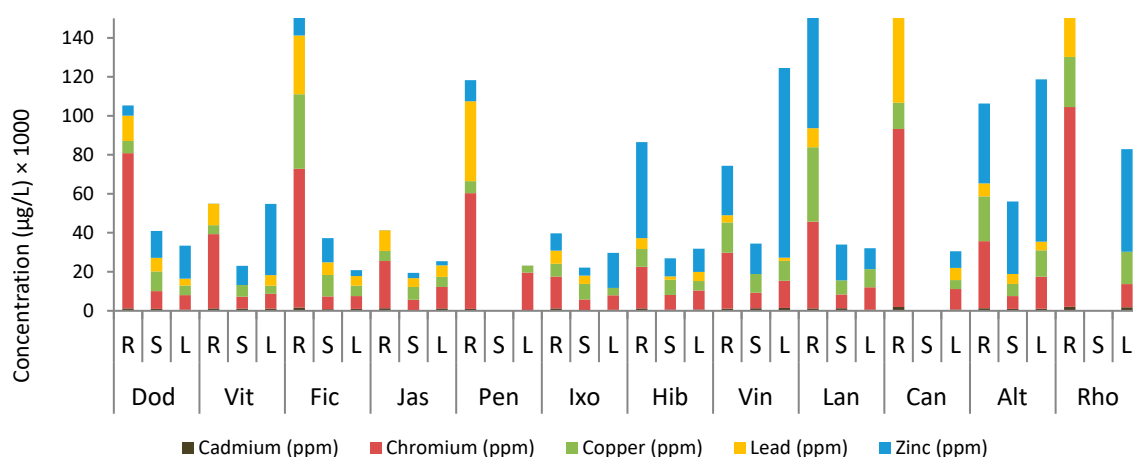

Figure 1. Average concentration of heavy metals in effluent water samples at the beginning (l, before irrigation with synthetic greywater) and at the end (F, after irrigation with synthetic greywater) of the experiment (please see Table S1 for the data).

3.2. Heavy Metals in Plant Samples

Figures 2 and 3 show the concentration of heavy metals in plant samples (roots, shoots and leaves) before and after greywater irrigation, respectively. It is evident that the concentrations of heavy metals in plant samples were significantly higher than those in the effluent water samples (Figure 1). At the beginning of the experiment (before greywater irrigation, Figure 2), the plants *Rhoeo discolor*, *Alternanthera ficondea* and *Lantana camara* had the highest concentration of metals, whereas the plants *Jasmine sambac* and *Pennisetum seateceum* exhibited the lowest concentration. The metals in the plant samples were mostly in the roots and leaves, whereas shoot samples had the lowest concentration in most of the monitored species. In most of the plants, Cr, Cu and Zn were present, whereas Pb was only observed in the plants *Pennisetum seateceum* (root) and *Canna indica* (leaves). Among all the plant species, concentration of Cr in the roots was higher than that of the other monitored metals, whereas leaves had higher concentrations of Zn.
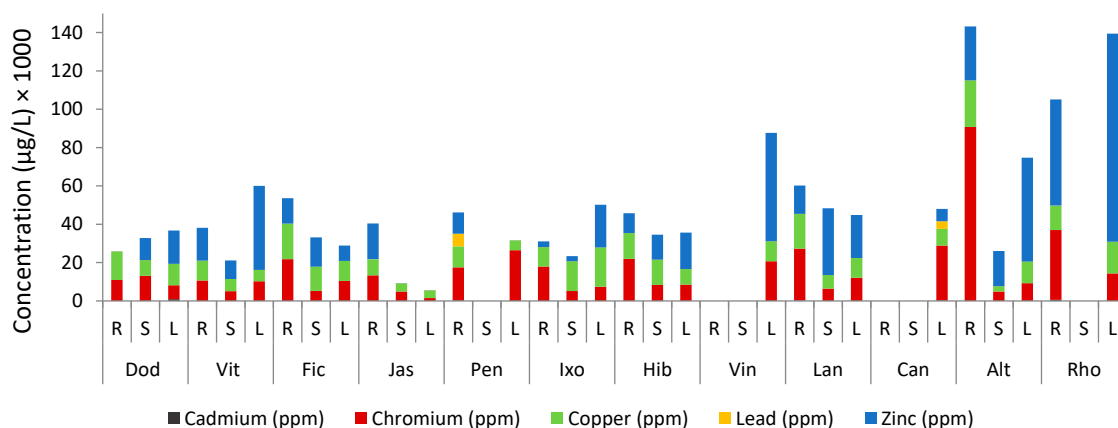
Figure 2. Average concentration of heavy metals in plant samples at the beginning of the experiment, before irrigation with synthetic greywater (R, S and L indicates roots, shoots and leaves, respectively) (please see Table S2 for the data).
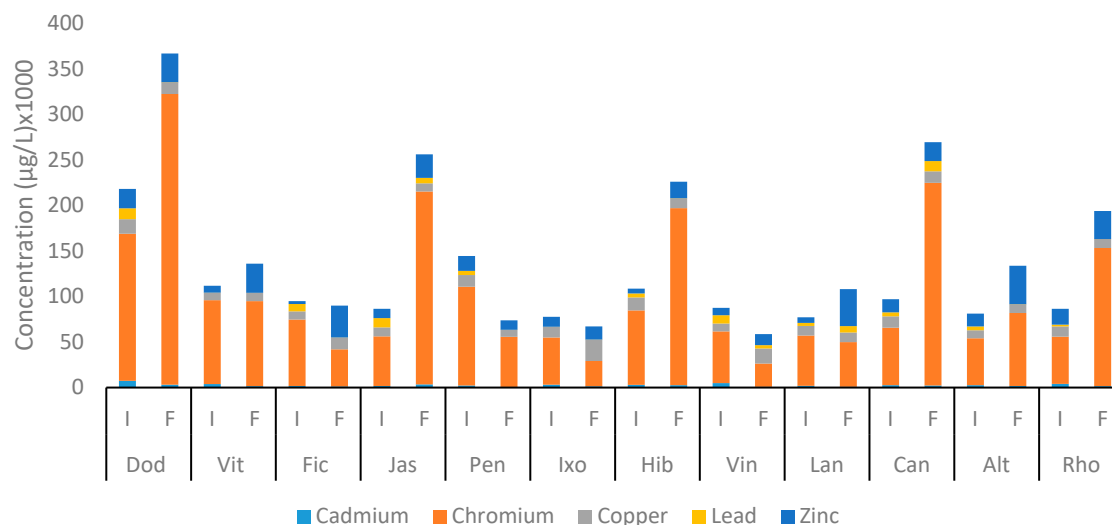

Figure 3. Average concentration of heavy metals in plant samples at the end of the experiment, after irrigation with synthetic greywater (R, S and L indicates roots, shoots and leaves, respectively) (please see Table S3 for the data).

Plants with the highest concentration of metals at the beginning (Figure 2) generally retained this status by the end of the experiment (Figure 3). The species *Jasmine sambac* had the lowest concentration of metals. The highest concentration of metals in all plant species remained in the roots and leaves, as observed at the beginning of the experiment. Interestingly, all plant species accumulated Pb in their roots by the end of the experimental period. Accumulation of Cd also increased by the end of the experiment. Overall, it can be seen that roots accumulated metals after irrigation with greywater.

3.3. Heavy Metals in Soil Samples

Figure 4 shows the concentration of heavy metals in soil samples used in the biofiltration columns, before and after irrigation with synthetic greywater. Among the five metals, Cr predominated in the soil samples both at the beginning and at the end of the experiment. The average concentration of total metals at the beginning was 106×10^3 µg/L and it was increased by approximately 50% to 165×10^3 µg/L by the end of the experiment. As shown in Figure 4 (I, before irrigation), soil samples from columns with the plant *Dodonaea viscose* contained the highest amount of metals (mostly Cr), followed by columns containing the plant *Pennisetum seateceum*. Although the concentrations of Cu and Zn were noticeable in soils for all columns, the presence of Cd was found minimal (7.3×10^3 µg/L for *Dodonaea viscose*, 4.8×10^3 µg/L for *Vinca rosea* and 3.9×10^3 µg/L for *Rhoeo discolor*), and Pb was not detected in columns with the plants *Vitex agnus* and *Ixora coccinaea*.

Figure 4. Average concentration of heavy metals in soil samples at the beginning (I, before irrigation with greywater) and at the end (F, after irrigation with greywater) of the experimental period (please see Table S4 for the data).

By the end of the experiment (Figure 4, after irrigation), metal concentrations in soil samples noticeably increased for the columns with the plants *Dodonaea viscose*, *Jasmine sambac*, *Hibiscus rosa-sinensis*, *Canna indica*, *Alternanthera ficondea* and *Rhoeo discolour*. In contrast, a decreased concentration was observed for columns with the plants *Ficus nitida*, *Pennisetum seateceum*, *Ixora coccinaea* and *Vinca rosea*, and their metal composition changed from being predominantly Cr at the beginning to a combination of Cr, Cu and Zn at the end. Irrespective of plant species, Zn concentration was increased after irrigation, whereas the concentration of Cu remained almost the same. Comparing the metal concentrations at the beginning and at the end (Figure 4), it can be seen that Cr concentration in *Dodonaea viscose* and *Hibiscus rosa-sinensis* almost doubled, and in *Jasmine sambac* and *Canna indica* it more than tripled by the end of the experiment. It can also be seen that Cd and Pb had the lowest average concentrations in soil samples for all columns at the beginning (3.2×10^3 µg/L and 5.193×10^3 µg/L, respectively) and at the end of the experiment as well (1.7×10^3 µg/L and 2.339×10^3 µg/L, respectively).

Soil type and composition both play a major role in retaining heavy metals in biofiltration systems [32]. Generally, fine-grained soil shows a higher capacity for heavy metal adsorption compared to coarse-grained soils [33]. The accumulation of metals in soil is governed by the adsorption process at the interfaces of inorganic colloids such as oxides, hydroxides and clay. The organic colloidal matters also serve as an interface for the adsorption process [31].

3.4. Distribution of Heavy Metals

3.4.1. Copper

Figure 5 shows the distribution of Cu in various components of the biofiltration systems before and after irrigation with synthetic greywater. It can be seen that Cu was present in all components of the biofiltration systems, which can be attributed to the fact that Cu generally occurs in soils [34] and in plants [35,36]. Plants adsorb Cu from fertilizers and pesticides [36] as well as from soils [35].

Before irrigation (Figure 5, I), plants accumulated Cu in the roots (42%), leaves (37%) and shoots (21%), and there was no detectable Cu in the soils. There was barely any difference between the plant species, except for those with no or less shoots (*Pennisetum seateceum* and *Rhoeo discolor*). The final distribution of Cu after irrigation (Figure 5, F) showed a significant change with respect to the accumulation of Cu in soil. Roots and soils were found to have the highest percentage of Cu (39% and 29%, respectively), followed by leaves (17%) and shoots (15%). Plants with no or short stem components, such as *Alternanthera ficondea* and *Ficus nitida*, exhibited a higher percentage of Cu in their

roots than the other components (soil and water), whereas for *Ixora coccinaea*, most Cu was in the soil. It is evident from Figure 5 that almost all of the Cu was captured by the system itself (soil and plants). Several previous studies also reported that biofiltration systems contain and store most of the influent Cu [7,8,14].
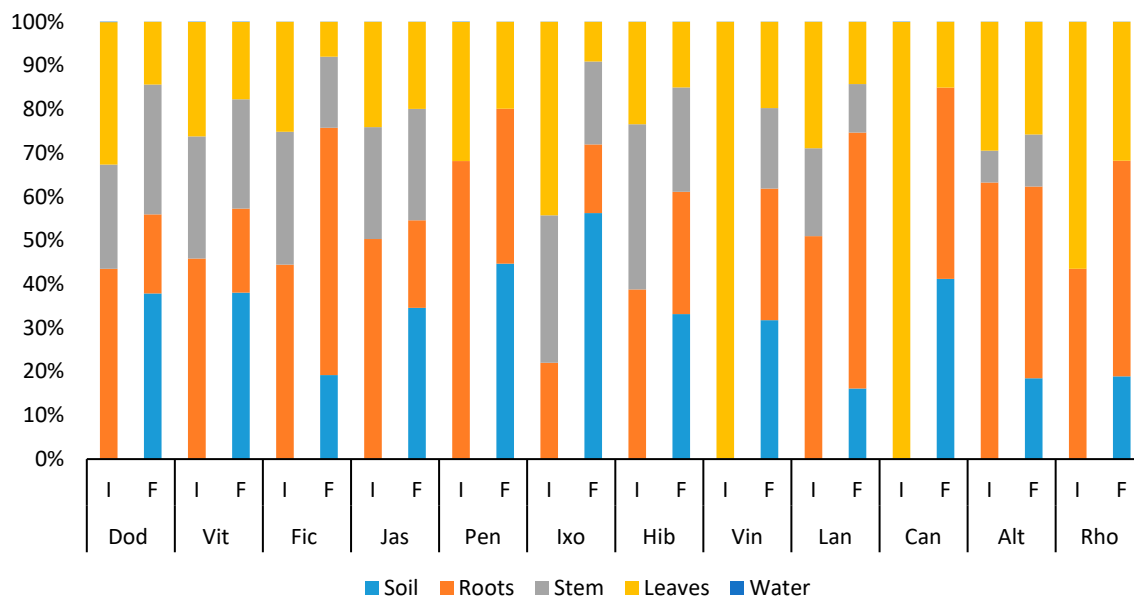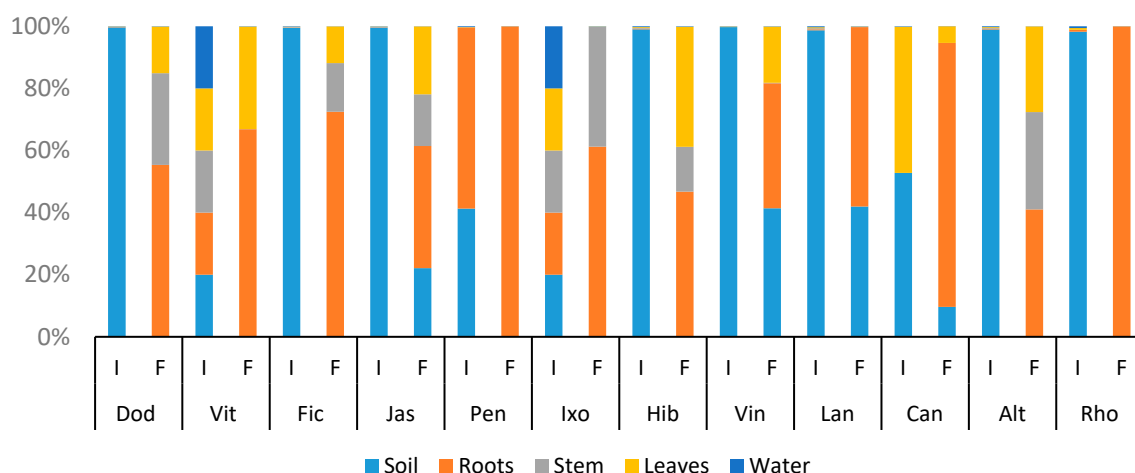

Figure 5. Distribution of Cu in the biofiltration system components at the beginning (I, before irrigation with greywater) and at the end (F, after irrigation with greywater) of the experiment.

Cu content in soils occurs in different forms apportioned between the dissolved and solid phases. The distribution within the different soil constituents is largely controlled by the amount of soil organic matter and the presence of manganese and iron oxides. Clay minerals and phosphates also play an important role [37]. A strong affinity between Cu and soil organic matter has been observed in previous studies, particularly when the organic fraction is high in relation to other metals [34].

3.4.2. Lead

Figure 6 shows the distribution of Pb in the biofiltration system components before and after irrigation with synthetic greywater. Before irrigation, Pb was found mostly adsorbed in soils for all columns except for the ones containing the species *Vitex agnus*, *Pennisetum seateceum*, *Ixora coccinaea* and *Canna indica*. In the columns containing the species *Vitex agnus* and *Ixora coccinaea*, Pb was almost equally distributed between soils, water, roots, shoots and leaves, whereas for the *Pennisetum seateceum* columns, Pb was mostly distributed in the roots (58%) and in the soils (42%), but for *Canna indica*, 47% was distributed in the leaves and the remaining 53% is in soils. By the end of the experiment, the Pb concentration in plants was higher than in the soils. Within the plants, the highest Pb concentration was in the roots, followed by shoots and leaves. The presence of organic matter in the biofiltration systems could be the reason for the migration of Pb concentration from soil samples at the beginning (reduced from 84% to less than 7%) to plant samples (increased from 16% to more than 93%) at the end. Bradl [31] also observed a similar phenomenon.
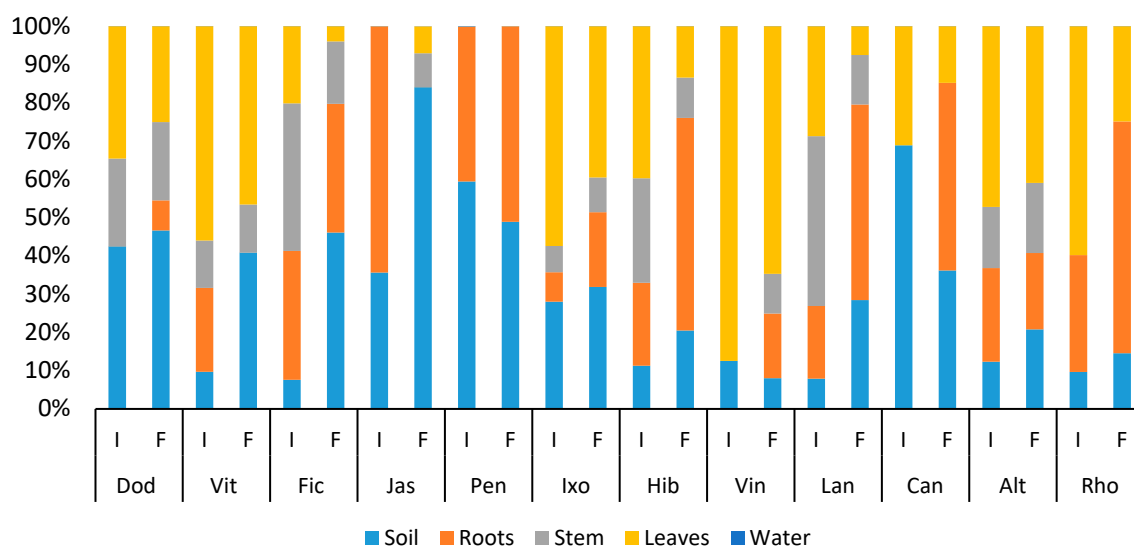
Figure 6. Distribution of Pb in the biofiltration components at the beginning (I, before irrigation with greywater) and at the end (F, after irrigation with greywater) of the experiment.

The reaction of Pb was influenced by adsorption, precipitation and the formation of complexes. Adsorption occurred at different solid phases, whereas precipitation affected the soluble and highly stable compounds. Stable complexes can form from the interaction of Pb with organic matter in the soil. At low pH, Pb undergoes and displays numerous hydrolysis reactions. A significant amount of $Pb(OH)_2$ is formed at higher pH levels (pH > 9), whereas $Pb(OH)^+$ predominates in the pH range between 6 and 10 [31]. Pb exhibits a biphasic behavior in its adsorption kinetics. At the beginning, the reaction is fast, which is likely caused by chemical reactions on the readily available surfaces. This is then followed by a slow reaction owing to diffusion to internal sites, possible formation of additional sorption sites and adsorption on top of low affinity sites that exhibit slow reaction rates [11]. Lead possesses a strong affinity to clays, peat, iron oxides and to normal soils [38], and its behavior is significantly influenced by the carbonate content of soil [31].

3.4.3. Zinc

Figure 7 shows how Zn is distributed in the biofiltration system components before and after irrigation with greywater. Before irrigation, more than 46% of Zn was found in the leaves, whereas the roots and soil contained only 22% and 16%, respectively. Shoots and water samples had the lowest percentage at both ends of the experiment for most of the plant species. Columns with the species *Jasmine sambac* and *Pennisetum seateceum* were alone in that soils and roots contained most of the Zn content at both ends of the experiment, unlike other plant species where leaves contained a significant percentage of Zn. Overall, at the end experiment (Figure 7, F), roots, leaves and soils together had more than 89% of the Zn concentration, whereas shoots and effluent water contained less than 11% and 0.01% of total Zn concentration, respectively. Previous studies [7–9,14,26] on biofiltration systems reported similar outcomes in terms of Zn removal and claimed that biofiltration systems are very effective in reducing Zn from influent waters. The concentration of Zn in soils is mainly governed by sorption, and this is affected by soil type, its organic content and pH. It is also affected by the clay mineral content, which increases the adsorption capacity of the soil [31].
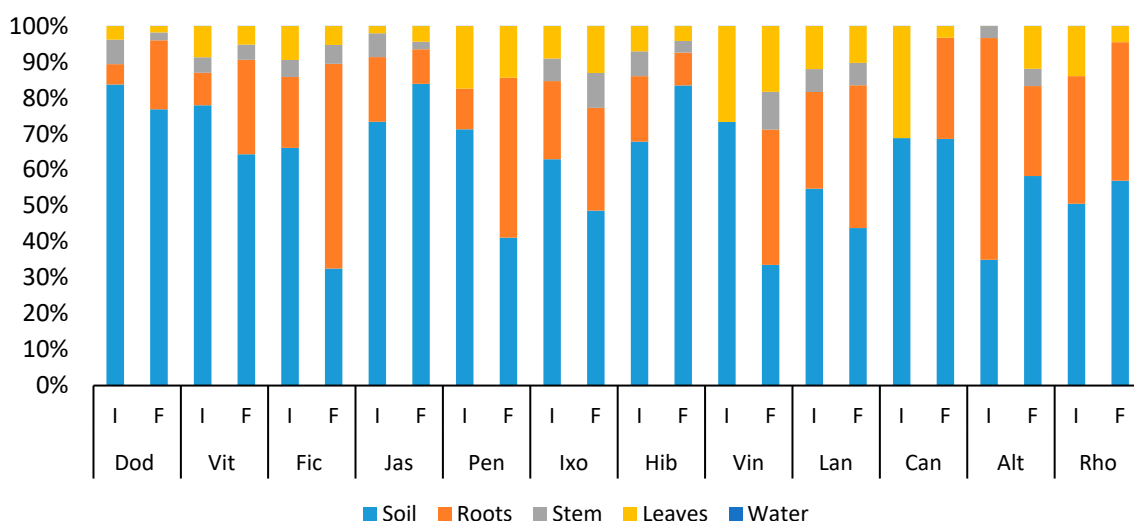
Figure 7. Distribution of Zn in the biofiltration components at the beginning (I, before irrigation with greywater) and at the end (F, after irrigation with greywater) of the experiment.

3.4.4. Chromium

Figure 8 shows the distribution of Cr in the biofiltration system before and after irrigation with greywater. This shows that the distribution of Cr remained almost unchanged in the soils (initially 65.3% and finally 64.6%), shoots (initially 3.9% and finally 2.7%) and in effluent water (initially 0.005% and finally 0.002%). However, Cr in the roots increased slightly at the end of the experiment (initially 19.4% and finally 26.9%). In contrast, Cr decreased slightly in the leaves at the end of the experiment (initially 11.4% and finally 5.8%). There are many factors that control the adsorption and precipitation behavior of Cr in soils. For example, soil pH increases the adsorption capacity. Other factors that affect the adsorption and precipitation of Cr in soil media are oxidation state, redox potential and the presence of competing ions and soil minerals [31].

Figure 8. Distribution of Cr in the biofiltration components at the beginning (I, before irrigation with greywater) and at the end (F, after irrigation with greywater) of the experiment.

3.4.5. Cadmium

The measured concentrations of Cd in the biofiltration system components were very low, and significant changes did not occur after greywater irrigation was applied to the columns. As shown

in Figure 1, before greywater irrigation, the effluent water samples from the columns of *Dodonaea viscose* and *Vitex agnus* had an average Cd concentration of 17.12 µg/L and 3.24 µg/L, respectively, whereas for all other columns, the concentrations were below 2 µg/L. After irrigation with greywater, the Cd concentration in effluent water samples from all the columns were below 4.4 µg/L (Figure 1, F). Negligible concentrations of Cd were observed in the soils and plants (roots, shoots and leaves) before and after irrigation with greywater (Figures 2–4, respectively). Cd occurs naturally in soils depending on the parent rocks from which the soils are formed. On average, the concentration varies from less than 0.1 to 0.3 mg/L for soils originating from igneous rocks to 0.3 to 11 mg/L for soils originating from sedimentary rocks [39]. At low concentrations, the reaction mechanism of Cd is mainly governed by adsorption [40] and more than 95% of this reaction occurs within 10 min of contact and an equilibrium is usually attained within an hour [41]. The solubility of Cd decreases with an increase in pH [42], and their adsorption in soils is largely affected by competing cations such as Ca and Zn.

This study explored the distribution of heavy metals in vegetative systems by providing a constant rate of greywater in all the 36 columns. The only variable considered was the plant. Previous studies have shown that metal distribution in vegetative systems is affected by some other factors such as inflow concentration, discharge ratio and recurrence interval on metals accumulation [43]. It was found that heavy metals are mostly accumulated in soils and roots. A recent study by Vatanpour et al. [44] exhibited the heavy metal contamination of food crops (cultivated rice) from irrigation water, which causes a health and ecological risk. To the best of our knowledge, metal accumulation in soil and their long-term impacts on the environment have not yet been investigated for urban green infrastructures.

4. Conclusions

This study explored the distribution of five heavy metals (Cd, Cr, Cu, Pb and Zn) in laboratory scale vegetative biofiltration columns before and after being irrigated with synthetic greywater. High concentrations of metals (thousands of µg/L) were measured in the plants, and the roots accumulated most of the heavy metals after irrigation with greywater. Of all the biofiltration components (water, soil and plants), soils accumulated the highest amount of heavy metals, both before and after irrigation with greywater. The outcomes of this study showed how biofiltration systems can be very effective in reducing heavy metals from influent wastewater. The vegetative system is becoming an integral part of urban landscape design, primarily because of its beneficial impact on stormwater source control and water quality improvement of receiving water bodies. This study showed that heavy metals are mostly accumulated in soils and plants; therefore, long-term sustainability of vegetative systems and their potential detrimental effects on the environment should be investigated. This study did not account for other influential parameters that affect heavy metal accumulation in vegetative systems such as inflow concentration, discharge ratio and recurrence interval on metal accumulation. Uncertainty analysis of data was not performed. Therefore, it is recommended that they be considered in future studies.

Nomenclature

Alt   *Alternanthera ficondea*
Can   *Canna indica*
Dod   *Dodonaea viscose*
Fic   *Ficus nitida*
Hib   *Hibiscus rosa-sinensis*
Ixo   *Ixora coccinaea*
Jas   *Jasmine sambac*
Lan   *Lantana camara*
Pen   *Pennisetum seateceum*
Rho   *Rhoeo discolor*
Vin   *Vinca rosea*
Vit   *Vitex agnus*

References

1. Woods-Ballard, B.; Kellagher, R.; Martin, P.; Jefferies, C.; Bray, R.; Shaffer, P. *The SUDS Manual*; CIRIA: London, UK, 2007; Volume 697.
2. Henderson, C.; Greenway, M.; Phillips, I. Sorption behaviour of nutrients in loamy-sand biofiltration media subject to different conditions (vegetation, enrichment and incubation time). In Proceedings of the 13th International Rainwater Catchment Systems Conference and 5th International Water Sensitive Urban Design Conference, Sydney, Australia, 21–23 August 2007.
3. Davis, A.P.; Shokouhian, M.; Sharma, H.; Minami, C. Water quality improvement through biofiltration media: Nitrogen and phosphorus removal. *Water Environ. Res.* **2006**, *78*, 284–293. [CrossRef] [PubMed]
4. Blick, S.A.; Kelly, F.; Skupien, J.J. *New Jersey Stormwater Best Management Practices Manual*; Chapter 9; New Jersey Department of Environmental Protection, Division of Watershed Management: Trenton, NJ, USA, 2004.
5. Davis, A.P.; Shokouhian, M.; Sharma, H.; Minami, C.; Winogradoff, D. Water quality improvement through biofiltration: Lead, copper, and zinc removal. *Water Environ. Res.* **2003**, *75*, 73–82. [CrossRef] [PubMed]
6. Geronimo, F.K.F.; Maniquiz-Redillas, M.C.; Kim, L.H. Fate and removal of nutrients in biofiltration systems. *Desalin. Water Treat.* **2015**, *53*, 3072–3079. [CrossRef]
7. Hatt, B.; Deletic, A.; Fletcher, T. Stormwater reuse: Designing biofiltration systems for reliable treatment. *Water Sci. Technol.* **2007**, *55*, 201–209. [CrossRef] [PubMed]
8. Davis, A.P. Field performance of biofiltration: Water quality. *Environ. Eng. Sci.* **2007**, *24*, 1048–1064. [CrossRef]
9. Trowsdale, S.A.; Simcock, R. Urban stormwater treatment using biofiltration. *J. Hydrol.* **2011**, *397*, 167–174. [CrossRef]
10. Li, H.; Davis, A.P. Heavy metal capture and accumulation in biofiltration media. *Environ. Sci. Technol.* **2008**, *42*, 5247–5253. [CrossRef]
11. Strawn, D.G.; Scheidegger, A.M.; Sparks, D.L. Kinetics and mechanisms of Pb (II) sorption and desorption at the aluminum oxide—Water interface. *Environ. Sci. Technol.* **1998**, *32*, 2596–2601. [CrossRef]
12. Elbana, T.A.; Ramadan, M.A.; Gaber, H.M.; Bahnassy, M.H.; Kishk, F.M.; Selim, H.M. Heavy metals accumulation and spatial distribution in long term wastewater irrigated soils. *J. Environ. Chem. Eng.* **2013**, *1*, 925–933. [CrossRef]
13. Turer, D.; Maynard, J.B.; Sansalone, J.J. Heavy metal contamination in soils of urban highways: Comparison between runoff and soil concentrations at Cincinnati, Ohio. *Water Air Soil Pollut.* **2001**, *132*, 293–314. [CrossRef]
14. Sun, X.; Davis, A.P. Heavy metal fates in laboratory biofiltration systems. *Chemosphere* **2007**, *66*, 1601–1609. [CrossRef] [PubMed]
15. Sarret, G.; Vangronsveld, J.; Manceau, A.; Musso, M.; D'Haen, J.; Menthonnex, J.; Hazemann, J. Accumulation forms of Zn andPb in Phaseolus vulgaris in the presence and Absence of EDTA. *Environ. Sci. Technol.* **2001**, *35*, 2854–2859. [CrossRef] [PubMed]

16. Madyiwa, S.; Chimbari, M.J.; Schutte, C.F.; Nyamangara, N. Greenhouse studies on the phyto-extraction capacity of Cynodont Nlemfuensis for lead and cadmium under irrigation with treated wastewater. *Phys. Chem. Earth* **2003**, *28*, 859–867. [CrossRef]

17. Kim, I.S.; Kang, K.H.; Perry, J.G. Investigation of heavy metal accumulation in polygonum thunbergii for phytoextraction. *Environ. Pollut.* **2003**, *126*, 235–243. [CrossRef]

18. Poschenrieder, C.; Bech, J.; Llugany, M.; Pace, A.; Fenés, E.; Barceló, J. Copper in plant species in a copper gradient in Catalonia (North East Spain) and their potential for phytoremediation. *Plant Soil* **2001**, *230*, 247–256. [CrossRef]

19. Chowdhury, R.K. Greywater reuse through a biofiltration system prototype in the arid region. *Water Sci. Technol.* **2015**, *72*, 2201–2211. [CrossRef]

20. Chowdhury, R.; Abaya, J.; Ksiksi, T.; Mohamed, M.M.A. Removal of heavy metals in vegetative greywater biofiltration columns. *J. Environ. Eng.* **2018**, *144*, 1–8. [CrossRef]

21. Fowdar, H.S.; Hatt, B.E.; Cresswell, T.; Harrison, J.J.; Cook, P.L.M.; Deletic, A. Phosphorus fate and dynamics in greywater biofiltration systems. *Environ. Sci. Technol.* **2017**, *51*, 2280–2287. [CrossRef]

22. Prathapar, S.A.; Jamrah, A.; Ahmed, M.; Al Adawi, S.; Al Sidairi, S.; Al Harassi, A. Overcoming constraints in treated greywater reuse in Oman. *Desalination* **2005**, *186*, 177–186. [CrossRef]

23. Eriksson, E.; Donner, E. Metals in greywater: Sources, presence and removal efficiencies. *Desalination* **2009**, *248*, 271–278. [CrossRef]

24. Rodda, N.; Salukazana, L.; Jackson, S.A.F.; Smith, M.T. Use of domestic greywater for small-scale irrigation of food crops: Effects on plants and soil. *Phys. Chem. Earth* **2011**, *36*, 1051–1062. [CrossRef]

25. Ghaitidak, D.M.; Yadav, K.D. Characteristics and treatment of greywater—A review. *Environ. Sci. Pollut. Res.* **2013**, *20*, 2795–2809. [CrossRef]

26. Turner, R.D.; Warne, M.S.J.; Dawes, L.A.; Vardy, S.; Will, G.D. Irrigated greywater in an urban sub-division as a potential source of metals to soil, groundwater and surface water. *J. Environ. Manag.* **2016**, *18*, 806–817. [CrossRef]

27. Al-Isawi, R.H.; Almuktar, S.A.; Scholz, M. Monitoring and assessment of treated river, rain, gully pot and grey waters for irrigation of Capsicum annuum. *Environ. Monit. Assess.* **2016**, *188*, 188–287. [CrossRef]

28. Diaper, C.; Toifl, M.; Storey, M. *CSIRO: Water for a Healthy Country National Research Flagship in Greywater Technology Testing Protocol*; CSIRO: Canberra, Australia, 2008.

29. Eriksson, E.; Auffarth, K.; Henze, M.; Ledin, A. Characteristics of grey wastewater. *Urban Water* **2002**, *4*, 85–104. [CrossRef]

30. EPA. *Hazardous Waste Test Methods/SW-846*; Environmental Protection Agency (EPA): Washington, DC, USA, 2014. Available online: https://www.epa.gov/hw-sw846 (accessed on 17 May 2016).

31. Bradl, H.B. Adsorption of heavy metal ions on soils and soils constituents. *J. Colloid Interface Sci.* **2004**, *277*, 1–18. [CrossRef]

32. Laurenson, G.; Laurenson, S.; Bolan, N.; Beecham, S.; Clark, I. The role of bioretention systems in the treatment of stormwater. *Adv. Agron.* **2013**, *120*, 223–274.

33. Crawford, R.J.; Harding, I.H.; Mainwaring, D.E. Adsorption and coprecipitation of single heavy metal ions onto the hydrated oxides of iron and chromium. *Langmuir* **1993**, *9*, 3050–3056. [CrossRef]

34. McGrath, S.P.; Sanders, J.R.; Shalaby, M.H. The effects of soil organic matter levels on soil solution concentrations and extractabilities of manganese, zinc and copper. *Geoderma* **1988**, *42*, 177–188. [CrossRef]

35. Deng, H.; Ye, Z.H.; Wong, M.H. Accumulation of lead, zinc, copper and cadmium by 12 wetland plant species thriving in metal-contaminated sites in China. *Environ. Pollut.* **2004**, *132*, 29–40. [CrossRef]

36. Khan, M.A.; Ahmad, I.; Rahman, I.U. Effect of environmental pollution on heavy metals content of Withania Somnifera. *J. Chin. Chem. Soc.* **2007**, *54*, 339–343. [CrossRef]

37. Jenne, E.A. Controls on Mn, Fe, Co, Ni, Cu, and Zn concentrations in soils and water: The significant role of hydrous Mn and Fe oxides. In *Trace Inorganics in Water*; Baker, R.A., Ed.; American Chemical Society: Washington, WA, USA, 1968; Volume 73, pp. 337–387.

38. McBride, M.; Sauve, S.; Hendershot, W. Solubility control of Cu, Zn, Cd and Pb in contaminated soils. *Eur. J. Soil Sci.* **1997**, *48*, 337–346. [CrossRef]

39. Kuo, S.; McNeal, B.L. Effects of pH and phosphate on cadmium sorption by a hydrous ferric oxide. *Soil Sci. Soc. Am. J.* **1984**, *48*, 1040–1044. [CrossRef]

40. Christensen, T.H. Cadmium soil sorption at low concentrations: I. Effect of time, cadmium load, pH, and calcium. *Water Air Soil Pollut.* **1984**, *21*, 105–114. [CrossRef]

41. Santillan-Medrano, J.; Jurinak, J.J. The chemistry of lead and cadmium in soil: Solid phase formation. *Soil Sci. Soc. Am. J.* **1975**, *39*, 851–856. [CrossRef]

42. Street, J.J.; Sabey, B.R.; Lindsay, W.L. Influence of pH, phosphorus, cadmium, sewage sludge, and incubation time on the solubility and plant uptake of cadmium. *J. Environ. Qual.* **1978**, *7*, 286–290. [CrossRef]

43. Li, Y.; Wen, M.; Li, J.; Chai, B.; Jiang, C. Reduction and accumulative characteristics of dissolved heavy metals in modified bioretention media. *Water* **2018**, *10*, 1488. [CrossRef]

44. Vatanpour, N.; Feizy, J.; Talouki, H.H.; Es'haghi, Z.; Scesi, L.; Malvandi, A.M. The high levels of heavy metal accumulation in cultivated rice from the Tajan river basin: Health and ecological risk assessment. *Chemosphere* **2020**, *245*, 125639. [CrossRef]