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SHORT TERM FATE OF HEAVY METALS IN THE GRAVEL BED HYDROPONICS WASTEWATER TREATMENT SYSTEM

The fate of five heavy metals was studied in a gravel bed hydroponics (GBH) wastewater treatment system. The study examined the removal of cadmium, chromium, copper, nickel, and zinc from an amended primary effluent. The removal of the metals from the effluent was determined as a function of time. A short term exposure, simulating a spill or shock load, of 6 to 8 hours was used. The concentration of the metals was determined in the associated sludge as well as in plant leaves, stems, and roots. Individually, good removals of chromium, cooper, nickel, and zinc were observed: > 70% average removal over 6 to 8-hour periods. Cadmium removal averaged 30% over an eight-hour experiment. When all five metals were spiked together, chromium removal was greater than 95%. Both copper and zinc were removed efficiently (> 90%) during the first three hours, and the removal efficiency dropped to about 70% by the sixth hour of exposure. Both nickel and copper were efficiently removed initially, but the removal dropped to 40% by the end of the experiment.

In this 12-meter long treatment system, the addition of heavy metals severely debilitated the crop plants in the first 2 to 3 meters. The remainder of the plants downstream were healthy and vigorous in appearance.

1. INTRODUCTION

Heavy metals are ubiquitous in natural waters in low concentrations. Domestic wastewater also contains variable amounts of different metals [1, 2]. Due to their refractory nature, their bioaccumulation in the food web and potential toxicity, heavy metals may have an adverse impact on the environment.

As a part of a larger project, we are treating a domestic primary effluent using gravel bed hydroponics (GBH). Gravel bed hydroponics is a process that uses plants to renovate wastewater and as a method of producing plant biomass (fig. 1). Micronutrient cations needed by these crops may be metals such as iron and

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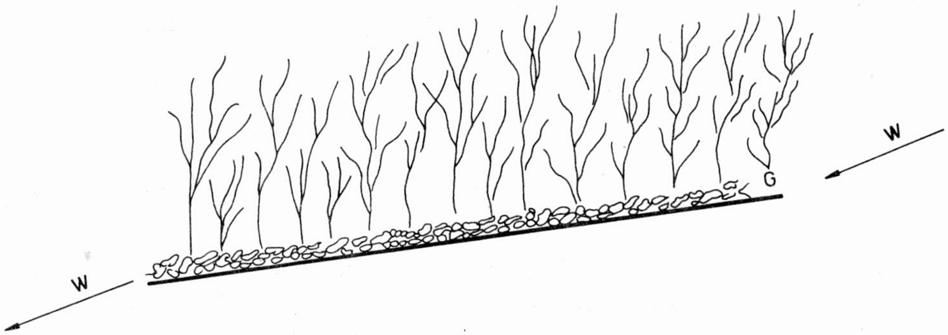

Fig. 1. Diagram of gravel bed hydroponics (GBH). Wastewater (W) flows into the top of the watertight channel, through the gravel (G) containing the crop roots, and out the bottom to a collection system. The slope is $5^0/_0$. The width of the channel is 1 m, the length is 12 m, and the depth is 20 cm. Depth of the gravel is about 5 cm. The plants remove nutrients (e.g., N and P), provide a physical filter and microbial habitat. The gravel distributes the wastewater evenly among the crop roots, prevents channeling of the wastewater, provides more surface area for microorganisms, increases wastewater retention time, and decreases effluent variability. The NFT is similar, except that it contains no gravel and is better suited for use in glass houses.

manganese [3, 4]. On the other hand, some other metals such as mercury, cadmium, chromium, and lead are considered toxic to these plants [4–8].

Domestic waste, in general, contains low concentrations of heavy metals, however it is possible that occasionally high levels of heavy metals could enter the system. High levels of heavy metals could adversely affect the plants, decreasing the effectiveness in treatment of the wastewater and/or accumulate in the plant and therefore the plant could not be used for domestic animal feed. Large metal accumulations would also interfere with use of the plant for chemical feed stocks [8–13], i.e. fermentation for acids, alcohols, and methane.

Vascular aquatic plants, employing solar energy as the principal energy source, have been shown capable of absorption, translocation and/or metabolic use of heavy metals and trace organics. WOLVERTON concluded from laboratory-scale wastewater investigations that water hyacinths can remove a maximum of 0.50 mg of nickel and 0.67 mg of cadmium per gram (dry weight) plant material over a 24-hr period [9]. A maximum concentration of 0.176 mg of lead and 0.150 mg of mercury per gram of dry plant tissue by water hyacinths has also been reported by WOLVERTON [10]. During the same study, alligator weed removed a maximum of 0.101 mg of lead per gram of dry plant tissue over 24 hr and a maximum of 0.150 mg of mercury per gram over 6 hours.

A field study on the tertiary wastewater treatment by the application of vascular aquatic plants has been reported [14]. The results of this study indicated that vascular aquatic plant systems can effectively reduce the trace contaminant content of secondary effluent to very low levels with essentially no energy requirements other than the sun. The removal of heavy metals obtained by the batch screening study was 82.1% for arsenic, 98.8% for cadmium, 92.8% for mercury and 94.9% for selenium. On the other hand, water hyacinth showed highest removal of arsenic (12.5%) and cadmium (68.6%), while duckweed showed highest removals of mercury (70.5%) and selenium (11.0%).

The present study was conducted to investigate the fate of five heavy metals in the GBH treatment system, i.e., cadmium, chromium, copper, nickel, and zinc. This study examined the removal rate of these five metals, the uptake of these metals by sludge and the translocation and accumulation of these metals in the plant leaves, stems, and/or roots. Detailed descriptions of the GBH and allied processes can be found elsewhere [9–13].

2. MATERIALS AND METHODS

The wastewater was artificially enriched with five metals individually or in combination as their soluble salts: Cd^{+2}, Cr^{+3}, Cu^{+2}, Ni^{+2}, and Zn^{+2}. These five metals were selected because of their potential toxicity to plants and their advance environmental effect. Each metal was added to the influent as the soluble nitrate salt. A level of 5 mg/dm^3 of each metal was maintained as an initial concentration. Each experiment was conducted for a 6 to 8 hour period.

Metal removal rates and the uptake of each metal by sludge and the plants were determined. For this purpose, samples from the influent and the effluent wastewater were collected, acidified, and filtered through Whatman No. 4. filter paper. Sludge and plant samples were collected at the beginning, in the middle, and at the end of each experiment. These samples were collected in duplicate at three different sampling locations along the reactor (0.25 m, 6 m, and 12 m, from the top of the reactor). The plant samples were partitioned into samples of roots, stems, and leaves. Sludge and plant samples were oven dried at 105 °C for 24 hours. The plant samples were then ground-up in a blender. The sludge and plant samples were weighed and digested using nitric acid followed by a hydrogen peroxide reflux [5, 6, 15]. Metal concentrations were determined using an instrumention laboratory atomic absorption spectrometer, model 251.

Each result is the average of 10-sequential readings. As an instrument and procedure blank, double distilled water, that was digested using the procedures previously described, was used.

3. RESULTS AND DISCUSSION

3.1. CADMIUM

Cadmium was added to the influent of the GBH trap at a concentration of 5 mg/dm^3 for 8.5 hours. Figure 2 shows the percentage removal in the effluent over the 8.5 hour period. The initial removal was about 60% which dropped to 20%

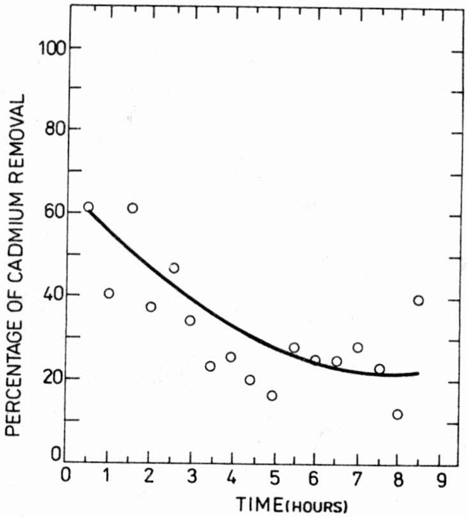

Fig. 2. Removal of cadmium in the Nutrient Film Wastewater Treatment process

at the end of the period. These removal rates do not appear to be sufficiently high to consider the GBH an effective process for treating a waste rich in cadmium.

Figure 3 shows the concentration of cadmium in the sludge at three sample points along the GBH tray. The GBH tray is 12 m long, therefore the top sample point was 0 m, the middle point was 6 m, and the bottom point was 12 m. These sample points were used consistently throughout this study.

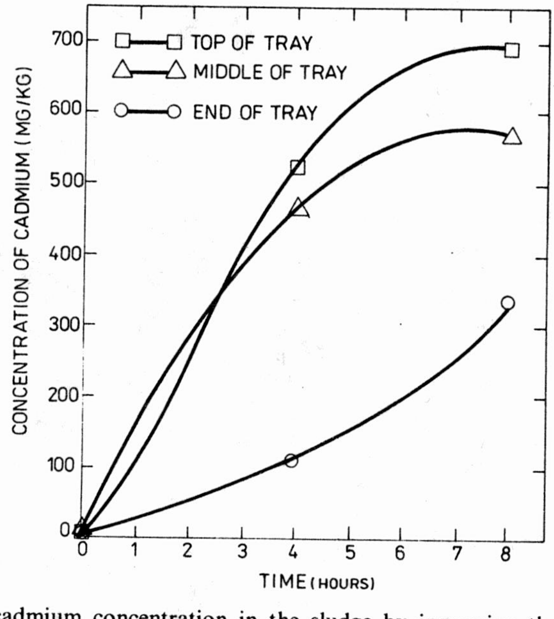

Fig. 3. Increasing of cadmium concentration in the sludge by increasing the exposure time along the tray

The cadmium concentration was approximately the same at all three sample oints at the beginning ($t = 0$) of the experiment, i.e. the background concentra- ons appeared to be the same throughout the tray. After four hours ($t = 4$) the top nd middle points showed an increase in the cadmium concentration, relative to he bottom point, which was much higher than at $t = 0$. In the 8-hr ($t = 8$) sample he concentration of the top and middle points, although higher than in the 4-hr ample, appeared not to have increased nearly as much as it did from $t = 0$ to $t = 4$. On the other hand, the bottom sample had increased substantially over the $t = 4$ sample. These results may indicate that the cadmium forms an equilibrium ith the sludge, and during a certain time the sludge becomes saturated with it (i.e. esults in a steady-state cadmium sludge concentration). As the system approaches quilibrium, the cadmium concentration reaching the lower sample point increases nd therefore the sludge–cadmium concentration also increases.

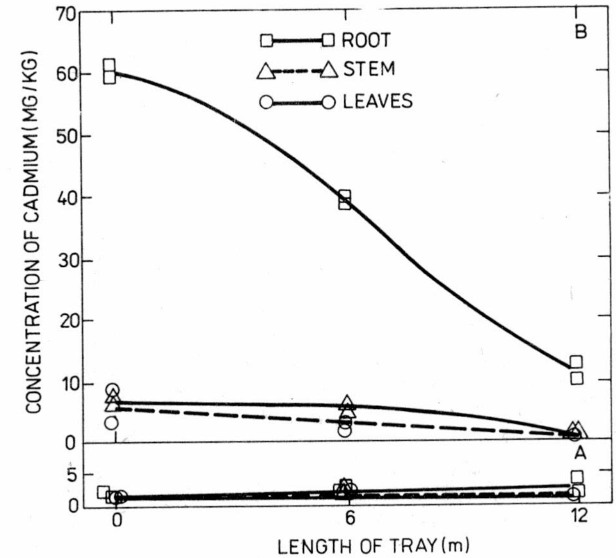

Fig. 4. Concentration of cadmium in plants

A – before adding $CdCl_2 \cdot 2\ 1/2 H_2O$. B – after the addition

Figure 4 shows the cadmium concentrations in the roots, stems, and leaves of ara grass (*Brachiaria mutica* = *Panicum purpurascens*), at the top, middle, and ottom sample points, before and after the addition of cadmium. The cadmium oncentration in all three plant parts was low prior to the addition of cadmium. After the experiment its concentration in the roots increased significantly at the) and 0.6 m sample points. At the 12 m sample point that concentration was higher than in the stem and leaf samples but was much lower than either at the 0 or 0.6 m sample points. The decrease in cadmium concentration in the roots from the 0 to 0.6 to 12 m sample is probably the result of a reduced aqueous cadmium concentration in the tray. This is consistent with the results obtained in the sludge–cadmium concentrations, which showed a similar decrease with increasing length.

3.2. CHROMIUM

Chromium was added to the influent in a concentration of 5 mg/dm^3 for 6.5 hours. In excess of 90% removal was achieved throughout the test period as shown in fig. 5. Figure 6 shows the concentration of chromium in the sludge. It

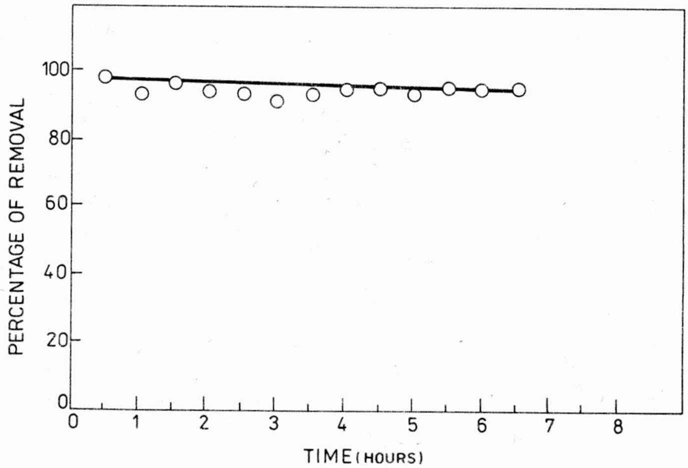

Fig. 5. Removal of chromium in the nutrient film wasterwater treatment process

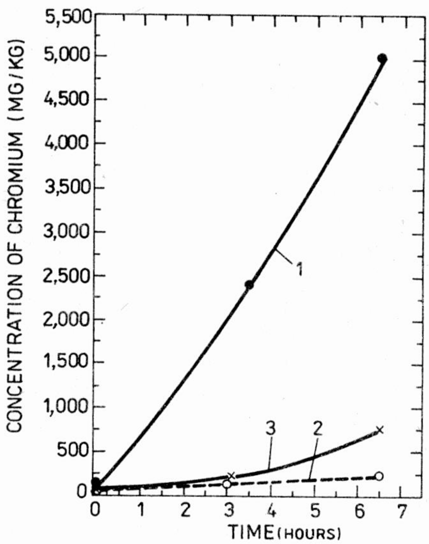

Fig. 6. Increasing of chromium concentration in the sludge by increasing the exposure time along the tray in the Nutrient Film Technique

1 – head of tray, 2 – middle of tray, 3 – end of tray

appears that one reason for the very good removal is the apparent strong complexation between the chromium and sludge. The chromium concentration in the sludge increased almost linearly with time at the 0 m sample point. Relatively speaking, very low chromium concentration was observed in either the middle or bottom sludge sample points.

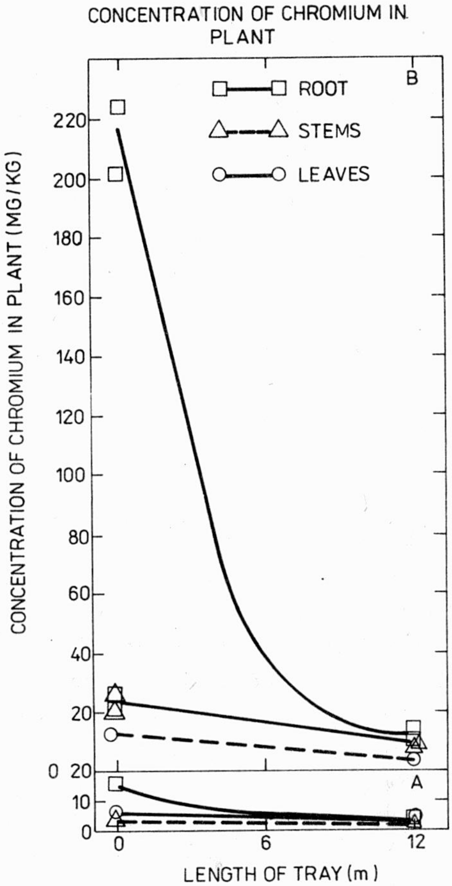

Fig. 7. Concentration of chromium in plants
A – before adding $Cr(NO_3)_3 \cdot 9H_2O$, B – after the addition

Figure 7 shows the concentration of chromium associated with the roots, stems, and leaves of para grass. The roots appear to concentrate most of the chromium from the wastewater. As was observed in the sludge, the majority had been removed at the initial (top) sample point, indicating a strong affinity (adsorption or absorption) for chromium.

3.3. COPPER

Copper was added to give an influent concentration of 5 mg/dm^3 over a five hour period. The influent and effluent concentrations as well as percentage removal of copper are given in fig. 8. This removal remained nearly constant at between 70 and 80% over the five hour test. It appears that the percentage removal would drop with an additional length of time.

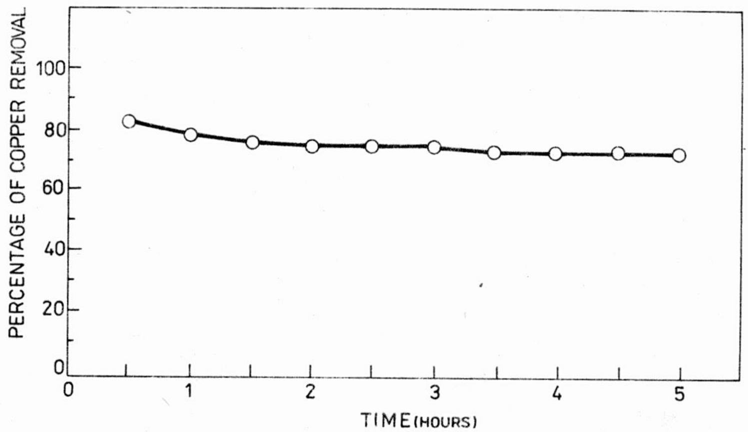

Fig. 8. Removal of copper in the Nutrient Film Wastewater Treatment process

Figure 9 shows the increase in copper concentration in the sludge at the three locations in the experimental tray. As expected, the copper concentration increased at each sample point with time and decreased slightly going from the influent to the middle sample point. The decrease in the sludge concentration at the bottom of the tray was much greater. These results probably reflect an equilibrium favouring the sludge (particulate) and therefore concentrating the copper at the top

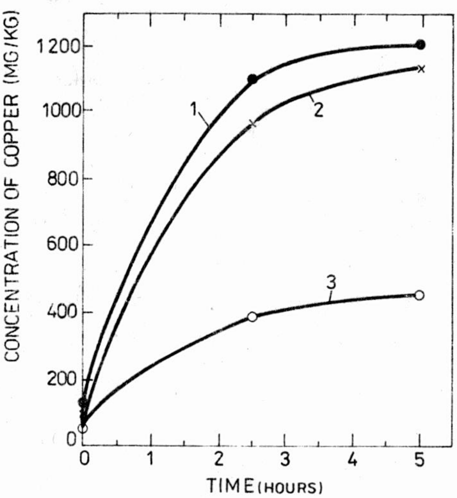

Fig. 9. Increasing of copper concentration in the sludge by increasing the exposure time along the tray

1 – head of tray, 2 – middle of tray, 3 – foot of tray

of the tray. Additional samples over several days would have provided a clearer picture of the ultimate fate of the copper associated with the sludge.

Figure 10 shows the copper concentration in the roots, stems and leaves of the para grass, before and after the experiment at the three sample locations in the tray. Very little increase was observed in the copper concentration associated with

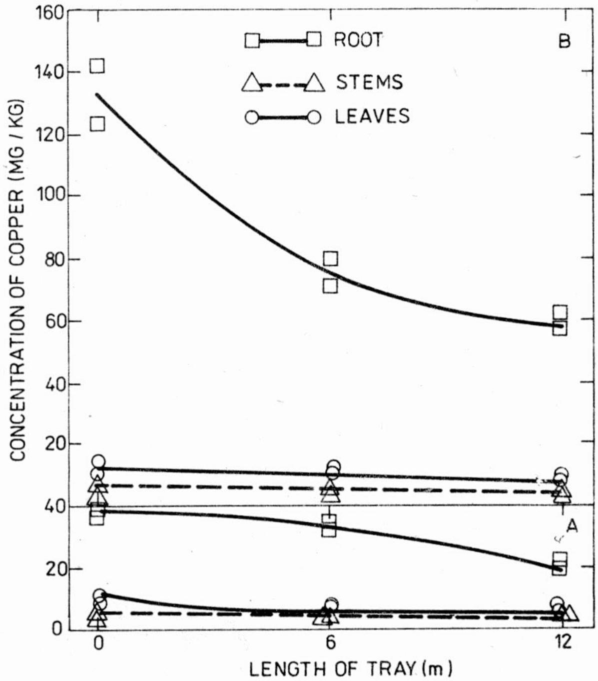

Fig. 10. Concentration of copper in plants

A – before adding $CuSO_4 \cdot 5H_2O$. B – after the addition

the stems and leaves. On the other hand, the roots showed an increase at every sample point. The increase was approximately threefold at each point, with an overall decrease in concentration from the top to the bottom of the tray. It appears that the background copper concentration, in the roots, decreased within the length of the tray. However, this is not statistically verified because of the widespread data points at the bottom of the tray. After the experiment the roots did show an overall increase in copper content, while its concentration decreased from the top of the tray to the bottom of the tray.

3.4. NICKEL

Nickel was added to the influent tray in the concentration of 5 mg/dm^3 for 7 hours. Figure 11 shows the removal of nickel in the effluent of the GBH over 7

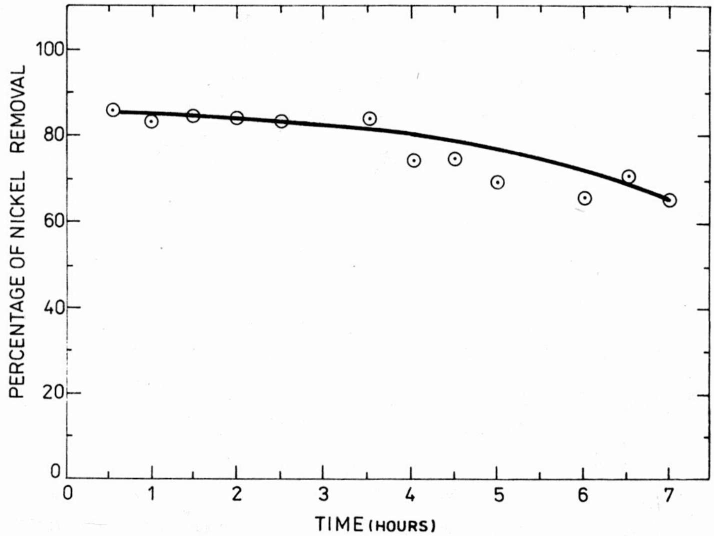

Fig. 11. Removal of nickel in the Nutrient Film Wastewater Treatment process

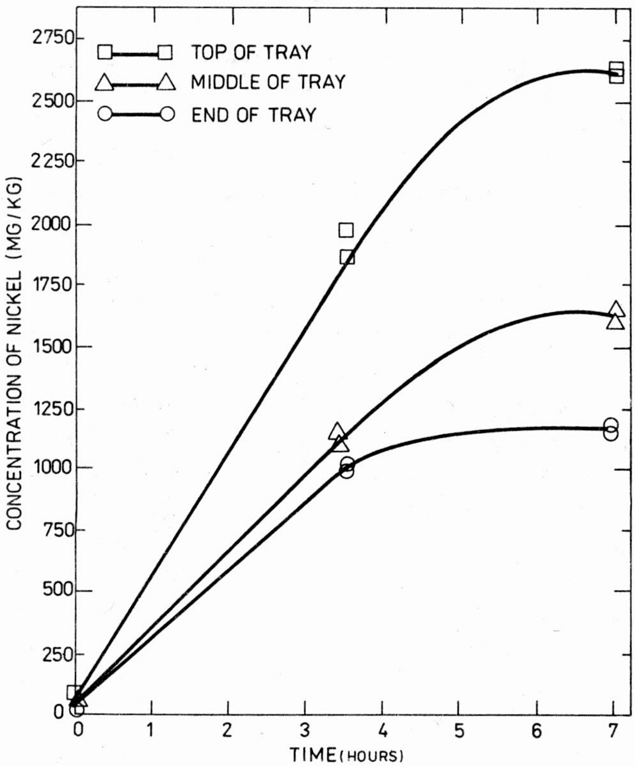

Fig. 12. Increasing of nickel concentration in the sludge by increasing the exposure time along the tray

hours. Initially, the removal was around 85% and this dropped to 65% removal at the end of the 7 hour test period. From the data it appears that the removal would have continued to drop had the experiment been continued for longer than 7 hours.

Figure 12 shows the nickel concentration in the sludge at the three sample points over the seven hour experiment. That concentration increased with time in all of the samples. It is evident that the increase between 3.5 and 7.0 hours was not nearly as great as from 0 to 3.5 hours. This could be explained if the system was approaching equilibrium.

Figure 13 shows the concentration of nickel in the plant roots, stems, and leaves before and after the experiment. As with the other metals, the roots appeared to accumulate most of nickel. The relatively higher concentration of nickel in the roots at the bottom sample point appears to follow the pattern observed in nickel concentrations in the sludge. Both of these observations indicate that nickel is not adsorbed by the sludge to the same extent as chromium. This also explains the poor removal of nickel when compared to chromium.

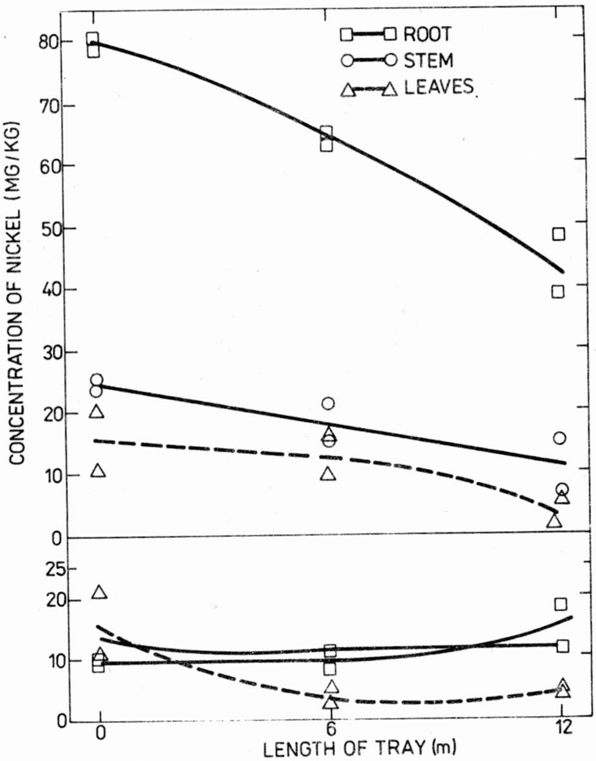

Fig. 13. Concentration of nickel in plants

A – before adding $Ni(NO_3)\cdot 6H_20$, B – after the addition

3.5. ZINC

Zinc was added to the influent in the concentration of 5 mg/dm^3 for 7.5 hour Figure 14 shows the concentration of zinc in the effluent of the GBH tray. It ca be seen that a steady increase was observed throughout the entire experiment, th is also demonstrated in the steadily decreasing percentage removal, from 90% t 65%.

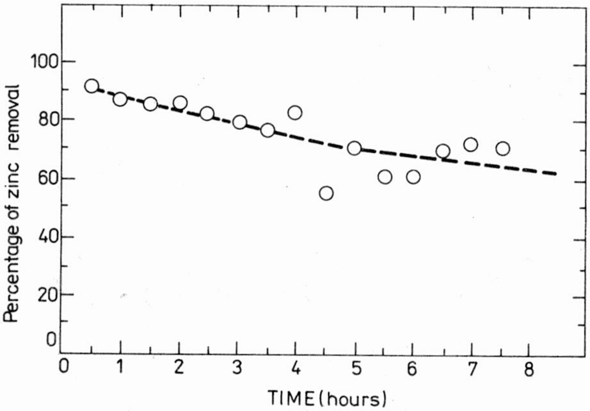

Fig. 14. Removal of zinc in the Nutrient Film Wastewater Treatment process

Figure 15 shows the concentration of zinc in the sludge, at the three samplin points. Figure 16 shows the distribution of zinc in the plant roots, stems, an leaves. Initially its concentration in the plant was higher than the concentration of other metals studied. It appears as though a concentration gradient existed i the plants before the addition of zinc in this experiment. This trend was no altered in the roots, where the zinc concentration went up at all sample points, bu did decrease with increasing length of the tray.

The data suggest that the adsorption of zinc in the sludge, being not as stron as for chromium, is similar to that of nickel. This has the effect of increasing it concentration in the water and giving rise to a linear decrease of zinc concentra tion in roots and small differences of zinc concentration in the sludge wit increasing tray length.

It appears from the data presented that one of the controlling factors in th removal of the five heavy metals studied is the removal in the sludge. The roo portion of the plants also concentrated the metals, the concentration is dependen on the metal. The mechanism of removal by the roots is not known and could b either adsorption or absorption. The roots were ashed prior to analyses, bu tightly bound (adsorbed) metal would probably not be removed.

No quantitative data exist on the mass per unit area of sludge, roots, stems o leaves necessary to complete a mass balance.

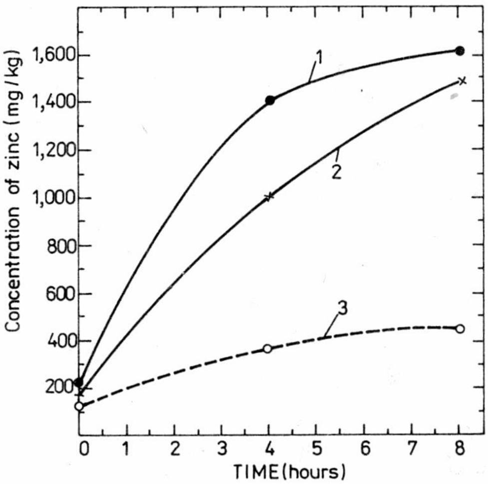

Fig. 15. Increasing of zinc concentration in the sludge by increasing the exposure time along the tray in the Nutrient Film Technique

1 – head of tray, 2 – middle of tray, 3 – end of tray

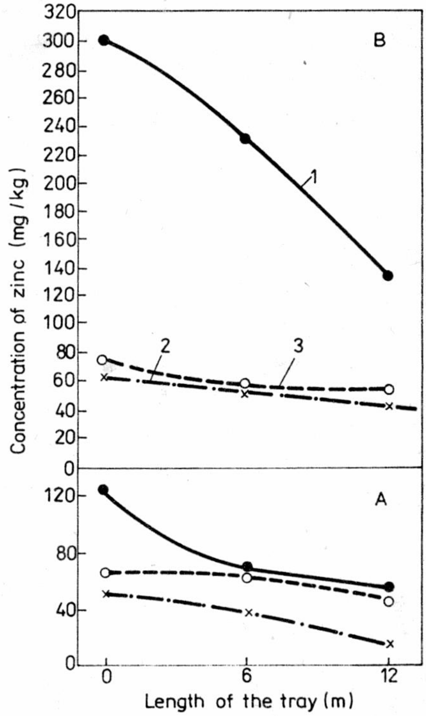

Fig. 16. Concentration of zinc in the plants

A – before adding $ZnSO_4 \cdot 7H_2O$, B – after the addition

1 – roots, 2 – stems, 3 – leaves

3.6. MIXTURE OF METALS

To simulate a spill and possibly a situation where more than one metal would be present, all five metals were added, in equivalent weight concentration (5 mg/dm^3) and their removal determined over 6.5 hours. Figure 17 shows the percentage removal of the five metals.

The results in fig. 17 seem to fall into these different groups. Chromium was effectively removed (> 95%) for the entire experiment. Both zinc and cadmium were effectively removed initially, but by the end of the experiment their removal was between 70 and 80%. Nickel and copper were not removed well, although nickel was initially removed (> 90%), both were removed in about 40% at the end of the experiment.

Chromium removal was found to be excellent in both experiments, when it was added individually or with all five metals. The removal of zinc was also about the same when added individually or in the mixture. These results indicate that

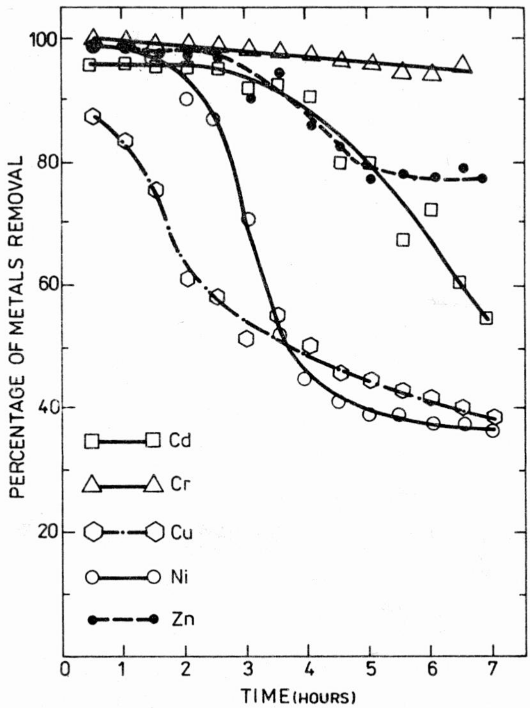

Fig. 17. Removal of cadmium, chromium, copper, nickel, and zinc in the Nutriemt Film Wastewater Treatment process

competition for adsorption sites did not adversely affect the removal of either chromium and zinc.

Cadmium, when added individually, was removed inefficiently, but when added in the mixture, the percentage removal increased. The explanation for this is not known.

Copper showed the opposite, it was removed rather uniformly ($\sim$ 70–80%) when added individually, but when added in the mixture, the percentage removal dropped to $\sim$ 40% by the end of the 6.5 hours. Nickel followed a similar pattern, showing very poor removal by the end of the experiment.

The reduced percentage removal of both copper and nickel in the mixture could be the result of competition for adsorption sites, in the sludge and roots, in the presence of the other metals.

Figure 18 shows the concentration of the five metals at the three sample points, in the GBH tray, during the eight hour experiment. Figures 19, 20 and 21 show the concentrations of the five metals, before and after the experiment, in the plant root, stem, and leaf samples.

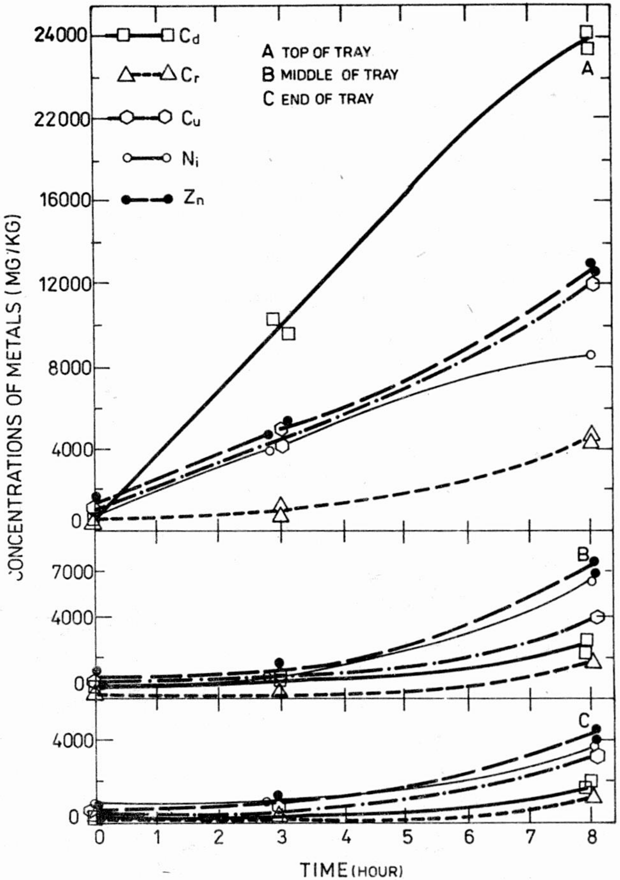

Fig. 18. Increasing of metals concentration in the sludge by increasing the exposure time along the tray in the Nutrient Film Technique

A – top of tray, B – middle of tray, C – end of tray

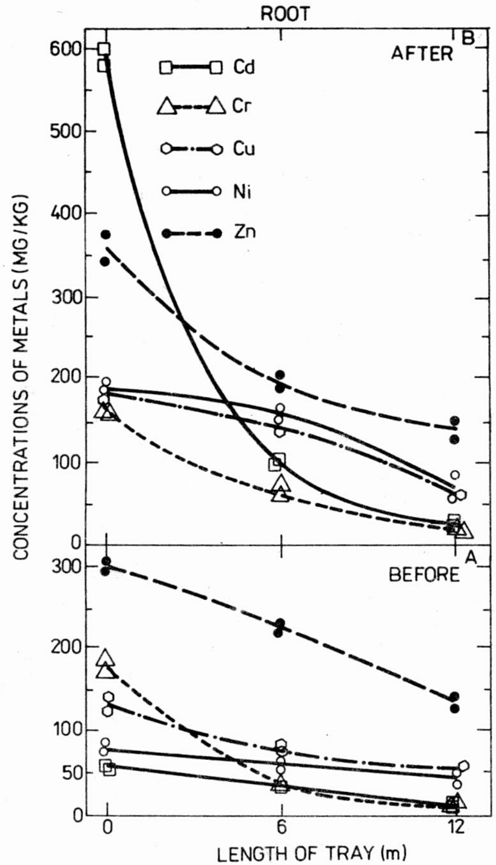

Fig. 19. Concentration of metals in the roots

A – before adding the metals, B – after the addition

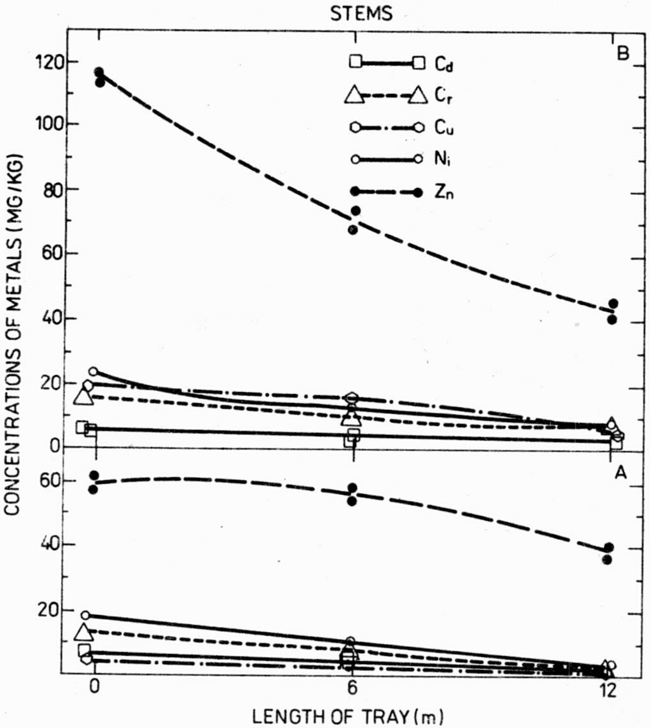

Fig. 20. Concentration of metals in the stems

A – before adding the metals, B – after the addition

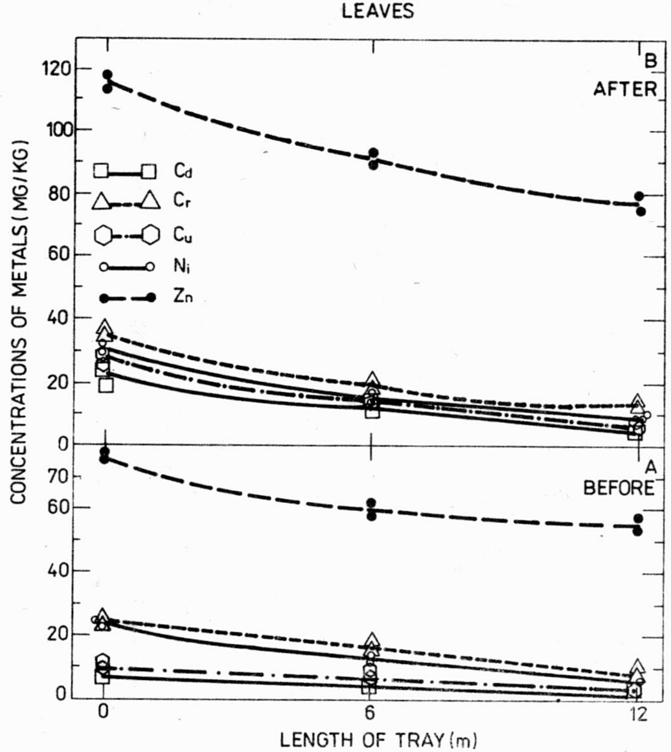

Fig. 21. Concentration of metals in the leaves

A – before adding the metals, B – after the addition

4. CONCLUSIONS

The present data showed that accumulation of five heavy metals in the sludge was the factor controlling their removal. The root portion of the plants also concentrated the metals much more than the stems and leaves. The concentration factor was dependent on the individual metal. The mechanism of removal by the root is not known and could be either adsorption or absorption.

The overall results indicated that the GBH wastewater treatment system can effectively reduce the concentration of heavy metals for a short term period. Removal of heavy metals (except for cadmium) obtained generally was greater than 95 to 65% with more than 95% for chromium trivalent. Removal rate of metals were in the order of:

$$Cr > Zn > Ni > Cu > Cd.$$

RECOMMENDATION FOR FUTURE STUDY

From the experiments conducted in this study it appears that additional studies are necessary. Foremost would be the extention of the multiple spiked heavy metal studies to be conducted over a long time period. A more thorough understanding of metal–organic and metal–inorganic interactions in the sludge would help to predict both short term and long term effects. The mechanisms of plant uptake are poorly understood and considerable work in this area would also provide data necessary to extend those studies.

ACKNOWLEDGEMENTS

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ZACHOWANIE SIĘ METALI CIĘŻKICH PODCZAS KRÓTKOTERMINOWYCH EKSPERYMENTÓW OCZYSZCZANIA ŚCIEKÓW W HYDROPONICZNYM SYSTEMIE O PODŁOŻU ŻWIROWYM

Zbadano zachowanie się pięciu metali ciężkich w hydroponicznym systemie oczyszczania ścieków o podłożu żwirowym. Badano usuwanie kadmu, chromu, miedzi, niklu i cynku ze wstępnie oczyszczonych ścieków. Określono stopień ich usunięcia w zależności od czasu ekspozycji. Stosowano krótkie czasy ekspozycji (6–8 godzin), symulując przelew lub obciążenie uderzeniowe. Mierzono stężenia metali w towarzyszącym osadzie oraz w liściach, łodygach i korzeniach roślin. Duże wydajności usuwania (70% po 6–8 godzinach) obserwowano dla chromu, miedzi, niklu i cynku. W przypadku kadmu przeciętna wydajność usuwania wynosiła 30% w czasie 8-godzinnego eksperymentu. Z mieszaniny wszystkich pięciu metali chrom usuwany był w $>95\%$, mieszanina miedzi i cynku natomiast usuwana była z wydajnością $>90\%$ w czasie pierwszych trzech godzin, a po sześciu godzinach ekspozycji spadała do $\sim 70\%$. Wydajność usuwania niklu i miedzi, początkowo dobra, spadała do 40% pod koniec eksperymentu.

Stosowany system hydroponiczny miał 12 m długości. Dodatek metali ciężkich do ścieków powodował wyraźne osłabienie uprawy tylko na pierwszych 2–3 metrach. Rośliny rosnące dalej były zdrowe i rozwijały się bujnie.

ПОВЕДЕНИЕ ТЯЖЁЛЫХ МЕТАЛЛОВ В КРАТКОВРЕМЕННЫХ ЭКСПЕРИМЕНТАХ ОЧИСТКИ СТОЧНЫХ ВОД В ГИДРОПОНИЧЕСКОЙ СИСТЕМЕ С ГРАВИЕВЫМ ОСНОВАНИЕМ

Исследовано поведение пяти тяжёлых металлов в гидропонической системе очистки сточных вод, использующей гравиевое основание. Были проведены исследования удаления кадмия, хрома, меди, никеля и цинка из предварительно очищенных сточных вод. Определено степень удаления этих металлов в функции времени экспозиции. Применены краткие времена экспозиции, от 6 до 8 часов, моделируя перелив или толчкообразную нагрузку. Измерялись концентрации металлов в сопровождающем осадке, а также в листьях, стеблях и кореньях растительных. Большие производительности удаления наблюдались особенно для хрома, меди, никеля и цинка. Они доходили до 70% после 6–8 часов. Для кадмия средняя производительность удаления составляла 30% во время восьмичасового эксперимента. Из смеси всех пяти металлов хром удалялся в свыше 95%. Смесь меди и цинка удалялась с большой производительностью ($>90\%$) во время первых трёх часов. После шести часов экспозиции эта производительность уменьшилась до ок. 70%. Производительность удаления никеля и меди была сначала хорошей, но в конце эксперимента уменьшилась до 40%.

Применяемая гидропоническая система имела 12 м. длины. Добавление тяжёлых металлов к сточным водам причинялось к отчётливому ослаблению культуры только на первых 2–3 метрах. Растения, растущие дальше были здоровыми и хорошо развивались.