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Removal of chloride salinity by aluminium, sodium and zinc compounds: impacts on water quality, survival and growth of *Hypophthalmichthys molitrix* Fry

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Abstract

The present study was conducted to remove chloride salinity from fish ponds because it is toxic to fish at concentrations of less than 0.05 mg/L. There are numerous methods currently employed to remove chloride from ponds. Use of chemicals is one of the alternatives for chloride removal. Experiment was conducted to remove chloride using Aluminium hydroxide, Sodium acetate, Aluminium nitrate and Zinc nitrate chemicals. The effect of various physico-chemical parameters of water has been investigated. The explanation of chloride removal by using these chemicals has also been supported by examining the survival and growth rate of fish. An increase in the values of physico-chemical parameters was observed except hardness which was decreased from its initial level. Zinc nitrate was found the most efficient in removing chlorides then Aluminium hydroxide and Aluminium nitrate while Sodium acetate the least efficient one. For fish survival and growth rate Aluminium hydroxide was found the most effective and Zinc nitrate the least effective one.

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Introduction

Chlorination is used in disinfecting and removing unwanted tastes and odours from drinking water. Sufficient chlorine is added to give a small excess for the destruction of pathogenic bacteria, but this excess can make municipal tap waters unsuitable for fish-keeping purposes. Chlorination is also used in textile and paper-pulp industries for bleaching and slimicidal purposes and in sewage treatment to reduce either odour, the density of ponding algae on filter beds, or the numbers of bacteria in effluents discharged to surface waters.

The amount of chlorine added to sewage is insufficient to oxidize the waste completely and there is no excess chlorine to impair the biological purposes essential for treatment, or the life in the waters to which the effluent is discharged. Chlorine is also added to cooling waters, and other industrial waste waters, to reduce or eliminate growths of algal or bacterial slimes in cooling towers and associated systems, and to swimming pools for the purpose of disinfection (Boyd, 1990).

Chlorine acts as a powerful oxidizing agent, and it is toxic to fish at concentrations of less than 0.05 mg/l. Chlorine as hypochlorous acid and chloramines is toxic to aquatic life. Concentration of chlorine greater than 8 µg HOCl/L could be harmful or lethal within four days to both salmonid and coarse fish.

The toxicity of chlorine to fish is increased by a reduction in the concentration of dissolved oxygen, and little changed up by increase in salinity up to 50% sea water (Alabaster and Lloyd, 1982).Chlorine can be removed from water by extended periods of aeration before use, or more rapidly by the addition of sodium thiosulphate at a rate of 7.0 mg/l for each 1 mg/L of chlorine. Sodium thiosulphate is not considered toxic to fish at concentrations required to remove chlorine from water (Boyd, 1990).

Since chlorine may react with thiocyanide to produce lethal concentrations of hydrogen cyanide and/or cyanogen chloride and concentrations of chlorine as low as 1 µg/L in the presence of phenols are likely to produce taints in the flesh of fish, an upper limit of less than 4 µg/L might be necessary in the presence of these other poisons. When fish exposed to chlorine solution becomes restless before losing equilibrium and dying (Alabaster and Lloyd, 1982). Enzymes within the cells contain sulphydryl groups essential for their activity and these become oxidized almost immediately by chlorine in both animals and plants, enzymatic activity being reversibly abolished because of the covalent bond formed.

This is the reason that once the equilibrium has been lost, the fish do not recover when placed in clean water; yellow perch tested at 10ºC appears to be a rare exception. Damage to gill epithelium has been reported by Penzes, 1971, Valenzuela, 1976, Bass *et al.*, 1977 and inferred by Rosenberger, 1971. However, Fobes, 1971found no effect on the respiration rate of isolated gill tissue excised from white sucker (*Catostomus commersoni*) exposed to 1mg/l chlorine, concluded that the gills were not the primary site of chlorine toxicity.

Acutely toxic concentrations of chlorine have increased the concentrations of various elements in the plasma of rainbow trout, especially that of potassium, and reduced the concentration of sodium (Zeitoun *et al.*, 1977).

In fathead minnow (*Pimephales promelas*) the acute toxicity of monochloramine has been ascribed to anoxia resulting from the production of methaemoglobin (Groethe and Eaton, 1975). Furthermore, young Coho salmon (*Oncorhynchus kisutch*) showed a slight increase in methaemoglobin in the blood and reduction in hemoglobin and a temporary increase in the percentage of circulating erythrocytes when exposed to chlorine at a concentration of 0.07 mg/l in sewage effluent diluted with sea water (Buckley, 1976) and mild to severe symptoms of hemolytic anemia after 12 weeks exposure to concentrations in the range 3-5 µg/l (Buckley, 1977). Although removal of chloride by calcined MgAl-CO_3 LDHs has been investigated by Kameda *et al.*, 2003, 2005 who employed ZnAl-NO_3 LDHs to remove chloride ion from aqueous solution in a batch mode by Liang (2006, 2009) and kinetic experiment was conducted to evaluate chloride removal using the Ultra-High Lime with Aluminum (UHLA) process (Abdel-Wahab and Batchelor, 2001, 2002, 2005; Abdel-Wahab *et al.*, 2005; Abdel-Wahaband Batchelor, 2006).

The removal of chloride salinity from aquatic media using Aluminium hydroxide, Sodium acetate, Aluminium nitrate and Zinc nitrate have not been reported yet. In the present work, the relative efficiency of these chemicals for chloride removal from aquatic media in the presence of silver carp fry has been evaluated.

The effect of various physico-chemical parameters has also been investigated. The explanation of chloride removal by chemicals has also been supported by examining the growth and survival rate of fish.

Materials and methods

Treatments& Chemical Reagents

The experiment was conducted in Chemistry Lab. Of Fisheries Research & Training Institute Manawan, Lahore. For the experiment 15 glass aquaria (30 liter/aquaria) were used.

There were five treatments with three replicates each. Aluminium hydroxide (Al $(OH)_3.9H_2O$), Sodium acetate (CH_3COONa), Aluminium nitrate (Al $(NO_3)_3$), and Zinc nitrate (Zn $(NO_3)_2.6H_2O$)were used as chemical reagents for the removal of chloride.

All the chemicals used in this study were of commercial grade and were used as received without purification. Variable under consideration was 'removal of chloride salinity from aquatic media' which was measured on alternate day basis.

Mode of Research &Analysis Work

First treatment T1 with its replicates T1R1, T1R2 and T1R3 was taken as control, having no chemical. Second, third, fourth and fifth treatments with triplicates were processed by utilizing chemicals. Physico-chemical parameters were sequenced to analyse the quality of water.

First readings for all parameters under consideration were interpreted before the use of proposed chemicals. 15 number of fish were numerated and added to each aquaria. Then 5g dose of the selected chemicals Aluminium hydroxide, Sodium acetate, Aluminium nitrate and zinc nitrate were supplemented to T2, T3, T4 and T5 treatments aquaria, respectively.

The survival rate of fish was examined and fluctuations in the physico-chemical parameters were observed in each treatment. Experiment was continued for 23 days and during this period, effect of chemicals on chlorides reduction was analysed.

Chloride was determined by titrating 25 ml of sample solution against Silver nitrate as a titrant using potassium chromate as an indicator from yellow colour till the end point that is brick red colour. Effectiveness of the chemicals in decreasing chloride level was evaluated. Effect of chemicals on the survival rate of fish was determined. Chloride level was checked out on daily basis. Average of triplicate readings was calculated. ANOVA statistical analysis on SPSS (version 22) was applied to find the significant differences for dissolved oxygen release by hydrogen peroxide (Steel *et al.,* 1996).

Results

Effect on Chlorides Removal

The data obtained from the removal of chloride salinity from aquatic media is presented in following Figures and Tables.

The results of the present study indicate the reductions in the chloride contents along with the alterations in physico-chemical parameters of pond water and effects on the fish survival and growth rates.

The chloride reductions trends are evident from data and graphs in Fig 1.

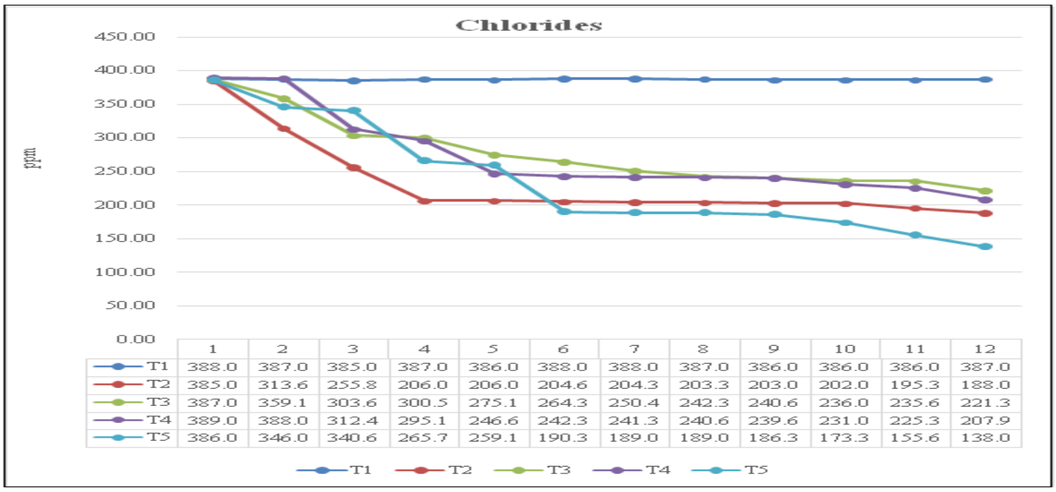

Fig. 1. Chlorides reduction trends in all the treatments.

The higher chloride reductions were found in Treatment 5 followed by T2, T4, T3 and T1 indicating that Zinc nitrate was better than Aluminium hydroxide which was again better than Aluminium nitrate, Sodium acetate and control respectively for chloride removal from the aquatic media.

Effect on Fish Survival Rates

The Fish Survival Rates are evident from data and graphs in Fig 2 showing the best treatment to be with T1 and T2 followed by T3, T4 and T5.

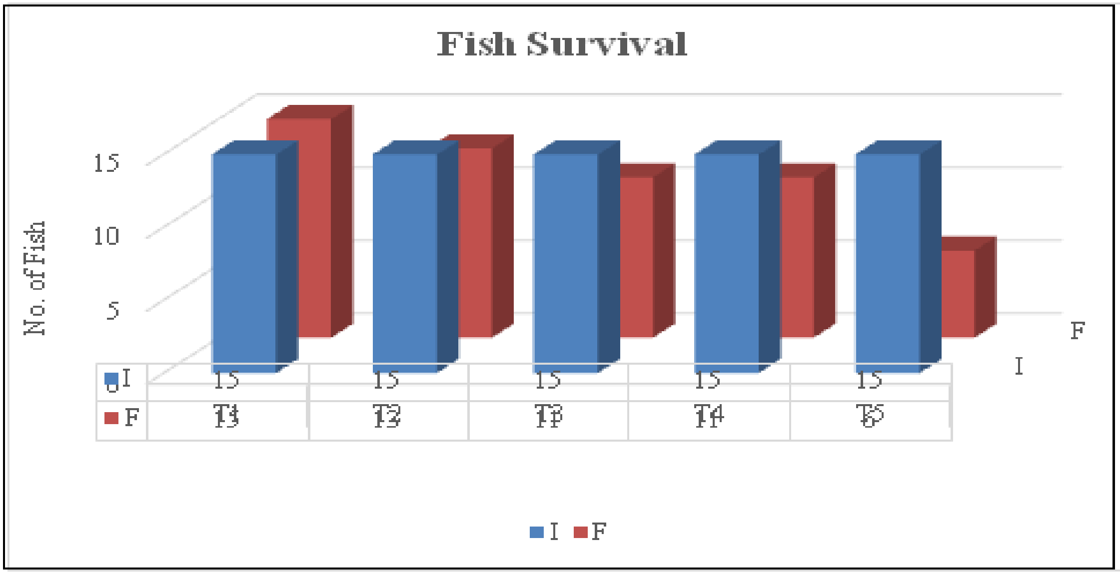

Fig. 2. Fish Survival Rates in all the treatments.

Effect on Fish weight

The Fish weight gains are evident from data and graphs in Fig 3 showing the best treatment to be with T2 followed by T3, T4, T1 and T5.

Effect on Temperature

The temperature was initially 28.17ºC then increased linearly to 30.57ºC and finally became 30.20ºC in T1 as showed by Fig 4.

The temperature was initially 28.03ºC then increased linearly to 30.77ºC and finally became 30.43ºC in T2 as showed by Fig 5.

The temperature was initially 28.33ºC then increased linearly to 30.93ºC. After gradual increase it showed curved behavior and then again decreased to 30.0ºC in T3 as showed by Fig 6.

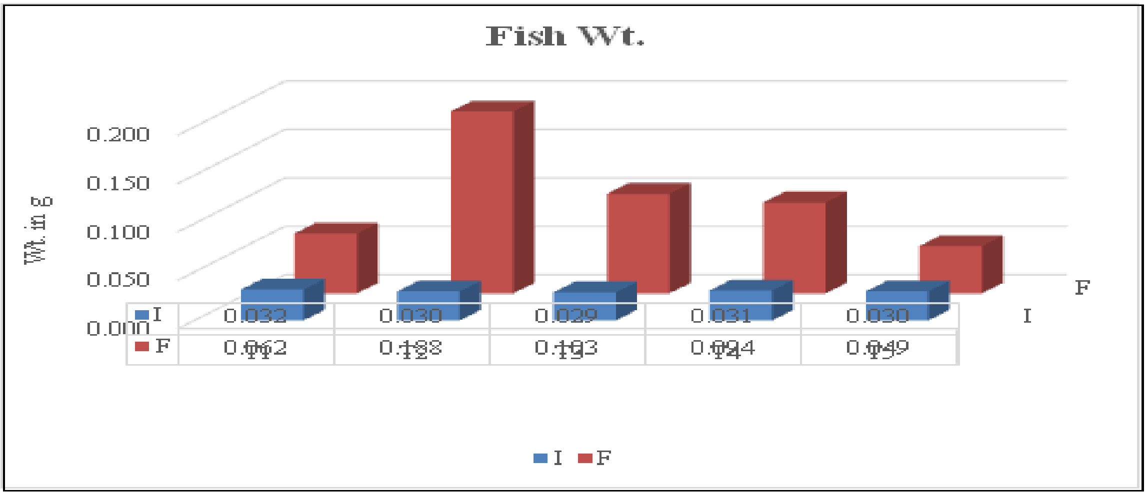

Fig. 3. Fish weight gain in all the treatments.

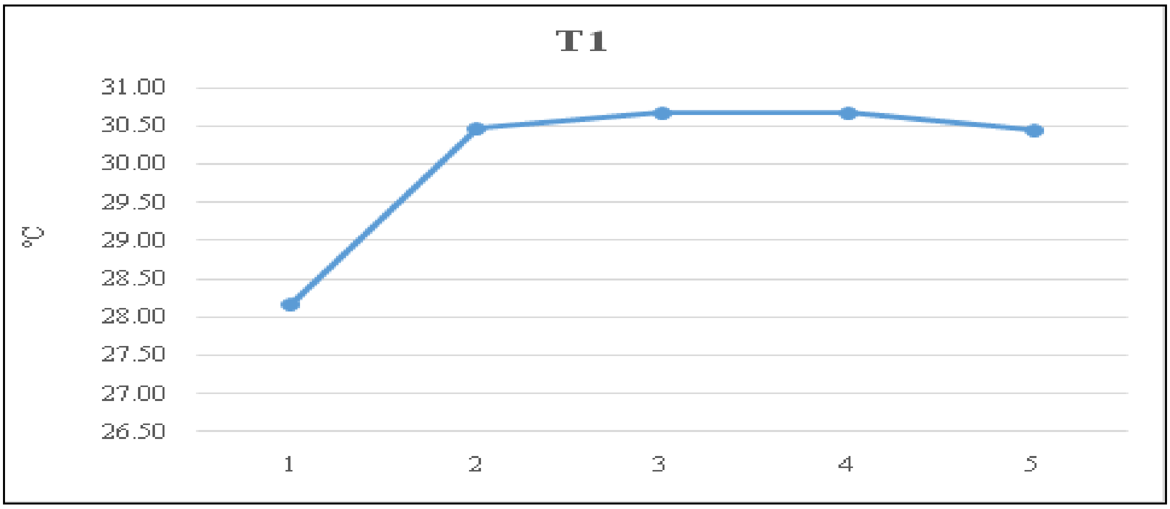

Fig. 4. Temperature profile of control (T1).

The temperature was initially 27.93°C then increased linearly to 30.57°C. After gradual increase it showed slightly curved behavior and then again decreased to 30.20°C in T4 as showed by Fig 7.

The temperature was initially 28.01°C, then increased to 30.57°C and showed horizontal line, then again decreased to making a V-shaped curve, then it again increased to 30.23°C as showed by Fig 8.

Effect on Alkalinity

The alkalinity values for all five treatments (i.e. T1, T2, T3, T4 and T5) were initially: 323.33 ppm, 314.00 ppm, 311.00 ppm, 316.33 ppm, 280.00 ppm respectively. After chemical treatments, values increased as such: 406.67, 394.67, 490.67, 328.00, 314.67 for T1, T2, T3 and T4 respectively as showed by Fig 9.

Effect on pH

The pH values showed by Fig 10for all five treatments (i.e. T1, T2, T3, T4 and T5) were initially: 8.52 ppm, 8.53 ppm, 8.52 ppm, 8.53 ppm, 8.52 respectively. After chemical treatment values increased as such: 8.98, 8.98, 9.04, 8.89, 8.86 for T1, T2, T3 and T4 respectively.

Effect on Conductivity

The conductivity values for all five treatments (i.e. T1, T2, T3, T4 and T5) were initially: 1161.33 μScm^{-1}, 1165.33 μScm^{-1}, 1280.67 μScm^{-1}, 1273.67 μScm^{-1}, 930.00 μScm^{-1} respectively. After chemical treatment values increased as such: 1231.33 μScm^{-1}, 1225.00 μScm^{-1}, 1450.67 μScm^{-1}, 1507.67 μScm^{-1}, 4843.33 μScm^{-1} for T1, T2, T3 and T4 respectively as showed by Fig 11.

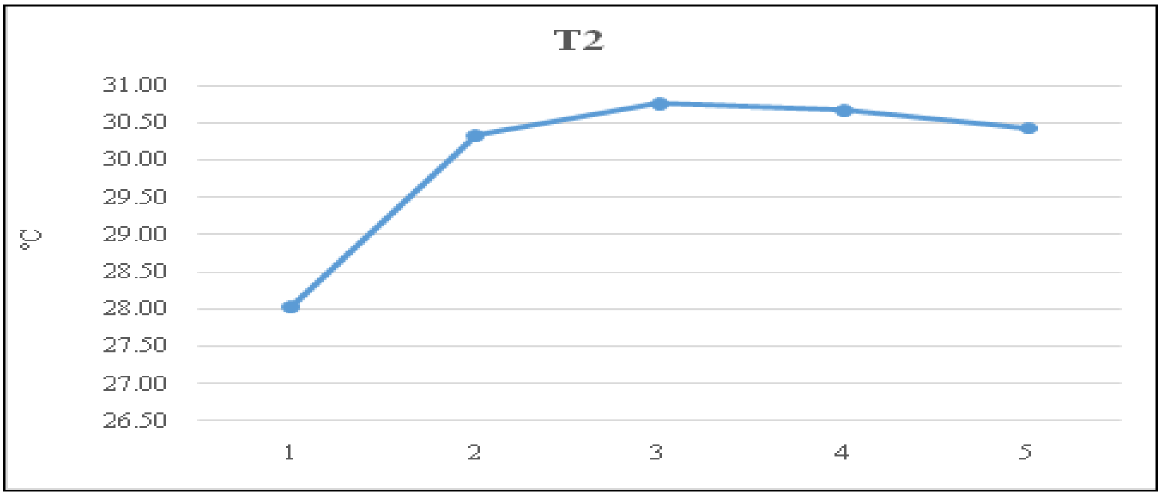

Fig. 5. Temperature profile of Aluminum hydroxide (T2).

Effect on Dissolved Oxygen

The dissolved oxygen values for all five treatments (i.e. T1, T2, T3, T4 and T5) were initially: 7.12 ppm, 7.22 ppm, 7.29 ppm, 7.37 ppm, 7.41 ppm respectively. After chemical treatment values increased as such: 7.79 ppm, 7.60 ppm, 7.51 ppm, 7.47 ppm, 7.53 ppm for T1, T2, T3 and T4 respectively as showed by Fig 12.

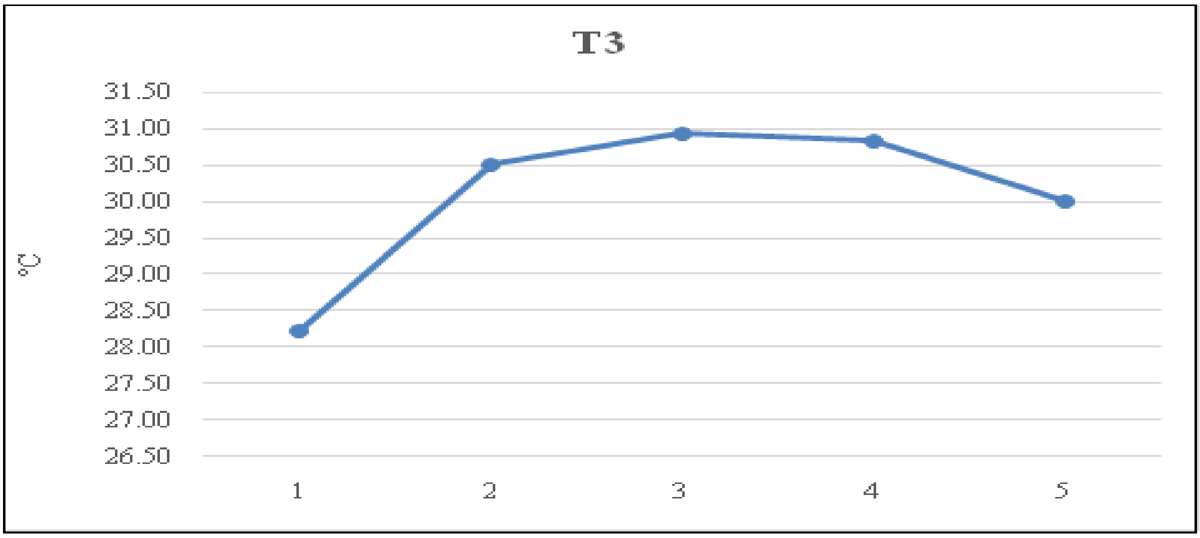

Fig. 6. Temperature profile of Sodium acetate (T3).

Effect on Hardness

The hardness values for all five treatments (i.e. T1, T2, T3, T4 and T5) were initially: 225, 245, 231, 221 and 227 ppm respectively. After chemical treatment values decreased as such: 200, 189, 163, 180 and 224 ppm for T1, T2, T3 and T4 respectively as showed by Fig 13.

Discussion

Chlorine is usually present at approximately 1mg/L in municipal water supplies as a result of chlorination. Fish succumb quite easily at these levels. Chlorine acts as a powerful oxidizing agent, and it is toxic to fish at concentrations of less than 0.05 mg/L. Water used for fish culture should not contain any residual chlorine to be considered safe. Chlorine can be removed from water by extended periods of aeration before use, or more rapidly by the addition of sodium thiosulphate at a rate of 7.0 mg/L for each 1 mg/L of chlorine. Sodium thiosulphate is not considered toxic to fish at concentrations required to remove chlorine from water.

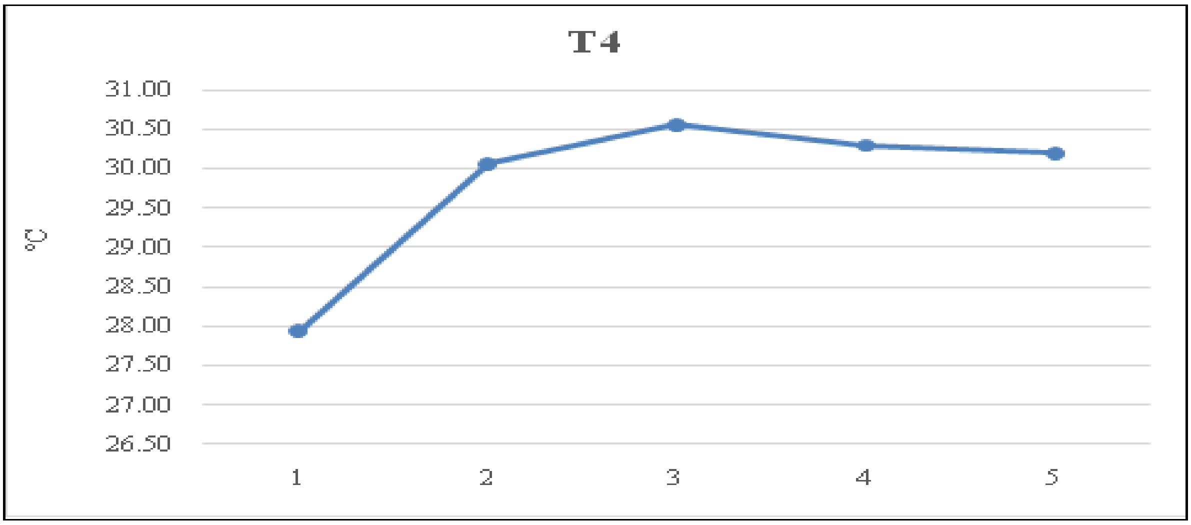

Fig. 7. Temperature profile of Aluminum nitrate (T4).

Chlorine as hypochlorous acid and chloramines is toxic to aquatic life. Concentration of chlorine greater than 8 µg HOCl/L could be harmful or lethal within four days to both salmonid and coarse fish.

There are various methods for the removal of chlorides but the removal of chloride salinity from fish ponds using chemicals like CH_3COONa, $Al(OH)_3(s)$, $Zn(NO_3)_2.6H2O$ and $Al(NO_3)_3.9H2O$ have not been reported yet. All the chemicals used in this study were of commercial grade. In the present work the relative efficiency of these chemicals for chloride removal from pond water has been evaluated. The effect of various physico-chemical parameters of water has been investigated. The explanation of chloride removal by using these chemicals has also been supported by examining the survival and growth rate of fish.

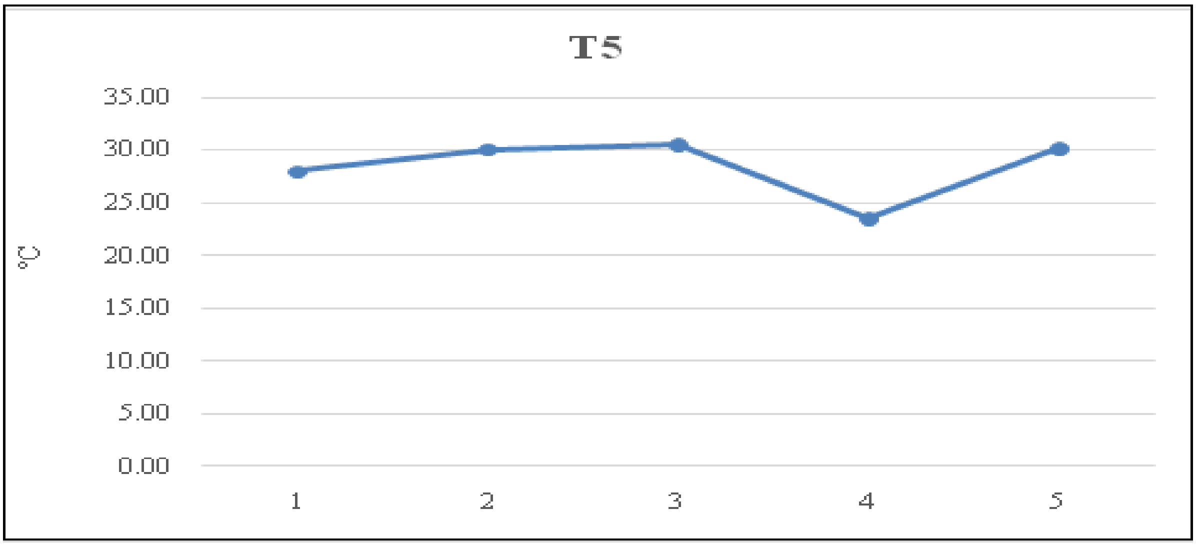

Fig. 8. Temperature profile of Zinc nitrate (T5).

Temperature of all the treatments increased initially and then decreased finally as shown in figures 4 to 8. But in T5,a V shaped curve was obtained i.e, after decreasing it again increased at the end.

An increasing trend was observed for all physico-chemical parameters of water except hardness.

Chlorides reduction trend in all the treatments was observed by examining the relative efficiency of chemicals.

Chlorides level in control T1 was almost same at both initial and final stages. Initially chlorides level was 388.0 and finally 387.00 ppm with a minute difference.

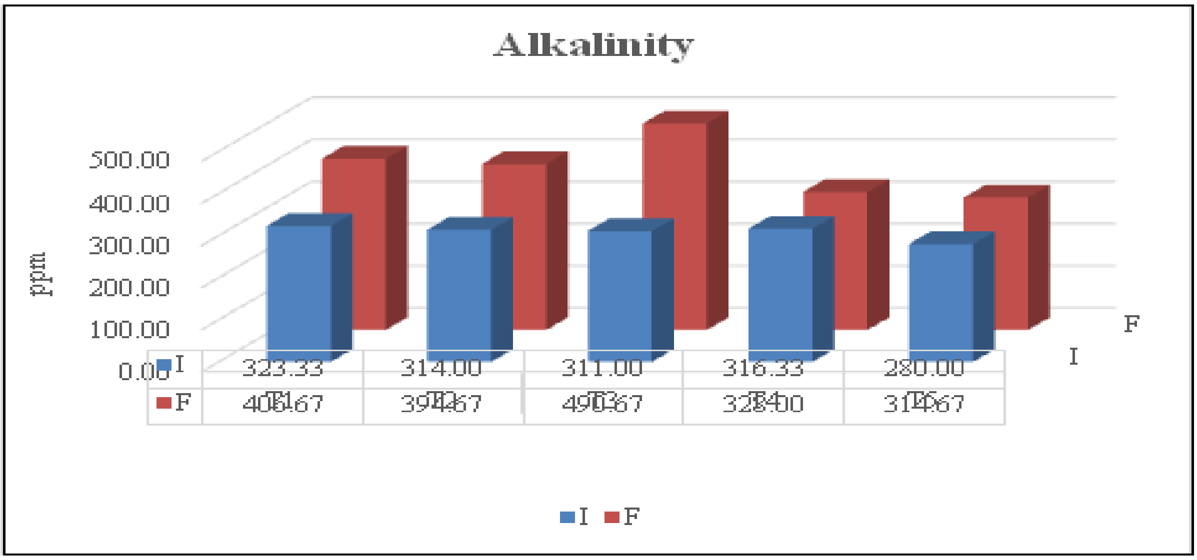

Fig. 9. Alkalinity variation in all the treatments.

In T2 treatment, Aluminium Hydroxide was used; the initial reading was 385.0, after adding chemical it decreased to 188.0 ppm. In T3 treatments Sodium acetate was used; the initial reading was 387.0, after adding chemical it decreased to 221.3 ppm. In T4 treatments; Aluminium Nitrate was added; initially chlorides level was 389.0 and finally decreased to 207.9 ppm. In T5 treatments Zinc Nitrate addition was added; the initial level of chlorides was 386.0 and finally it reached to 138.0 ppm.

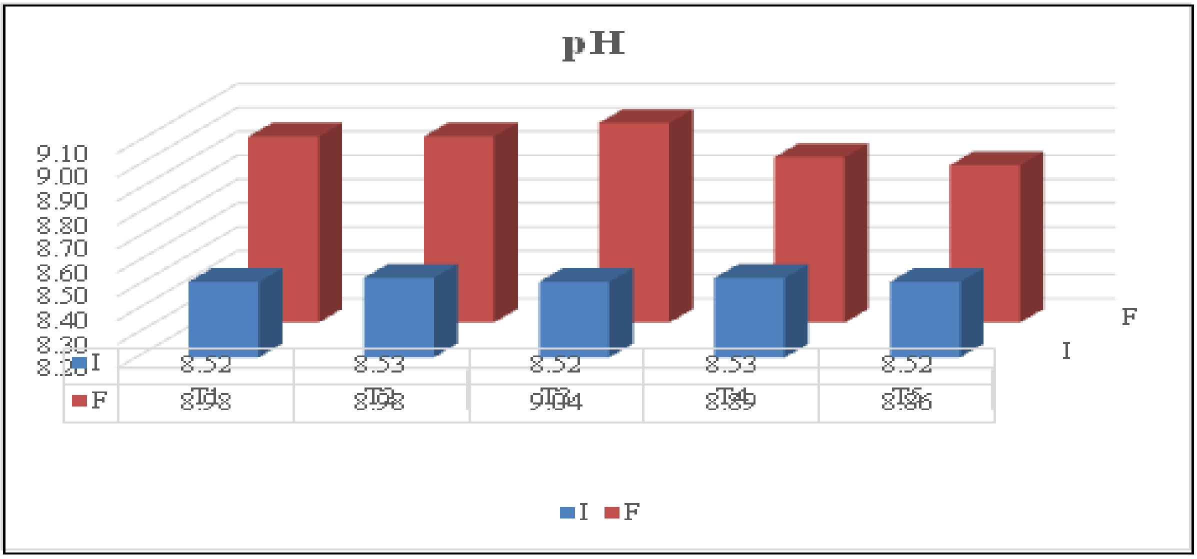

Fig. 10. pH variation in all the treatments.

By analyzing all the data available from the experiment, it was observed that Zinc Nitrate was the most efficient of all the chemicals in decreasing chlorides level. Aluminium Hydroxide was less efficient, then Aluminium Nitrate and then Sodium acetate having the least efficiency. The decreasing trend of the efficiency of chemicals on chlorides removal can be shown as:

Zinc nitrate > Aluminium hydroxide > Aluminium nitrate > Sodium acetate.

Effect of these proposed chemicals on survival rate of fish was also evaluated. In control there was no decrease in the no. of fish. In T2 treatments containing Aluminium Hydroxide fish number was decreased from 15 to 13. In T3 and T4 treatments containing Sodium acetate and Aluminium Nitrate respectively fish was decreased from 15 to 11. By analyzing all the data available from the experiment, it was observed that Aluminium Hydroxide was the most effective of all the chemicals for survival rate of fish.

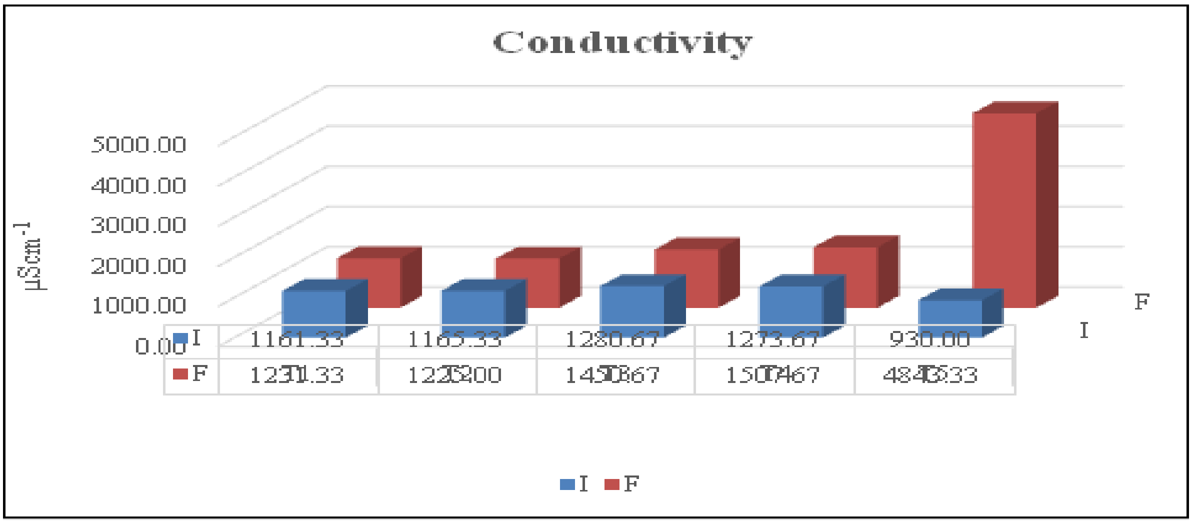

Fig. 11. Conductivity variation in all the treatments.

Sodium acetate was less efficient, then Aluminium Nitrate and then Zinc Nitrate having the least efficiency. The decreasing trend of the relative effectiveness of chemicals on fish survival rate can be shown as:

Aluminium hydroxide > Sodium acetate > Aluminium nitrate > Zinc nitrate.

Effect on fish weight gain in all the treatments was also evaluated. Within 23 days of experimental work fish gained weight. Initial and final readings of fish weight were noticed. In T1 treatments initial weight of fish was 0.032g and it increased to 0.062g at the end. In T2 treatments initial weight of fish was 0.030g which gained to 0.188g. In T3 treatments initial weight of fish was 0.029g that was increased to 0.103g. In T4 treatments Initial weight was 0.031g and finally gained 0.094g. In T5 treatments initial weight of fish was 0.030g and final weight was 0.049g. The decreasing efficiency of fish weight gain in all the treatments can be shown as:

Aluminium hydroxide > Sodium acetate > Aluminium nitrate > control > Zinc nitrate.

By analyzing all the data available from the experiment, it was observed that Aluminium Hydroxide was the most effective of all the chemicals for fish weight gain. Sodium acetate was less efficient, then Aluminium Nitrate and then Zinc Nitrate having the least efficiency.

Similarly, previous literature also supports our research through different chemical usage for chloride salinity removal as explained by Miller *et al.,* 2001 who used an optional pretreatment which removed chloride via oxidation to chlorine using sodium bismuthate, separating the sample aqueous and solid components for chloride removal and determining it through titration or by spectrophotometric method. Our research is also in agreement with the work of Liang *et al.,* 2009 who employed a chemical ZnAl-NO3 LDHs to remove chloride ion from aqueous solution in a batch mode through the high anion-exchange capacity of layered double hydroxides (LDHs) containing nitrate as the interlayer anion in the presence of appropriate anions.

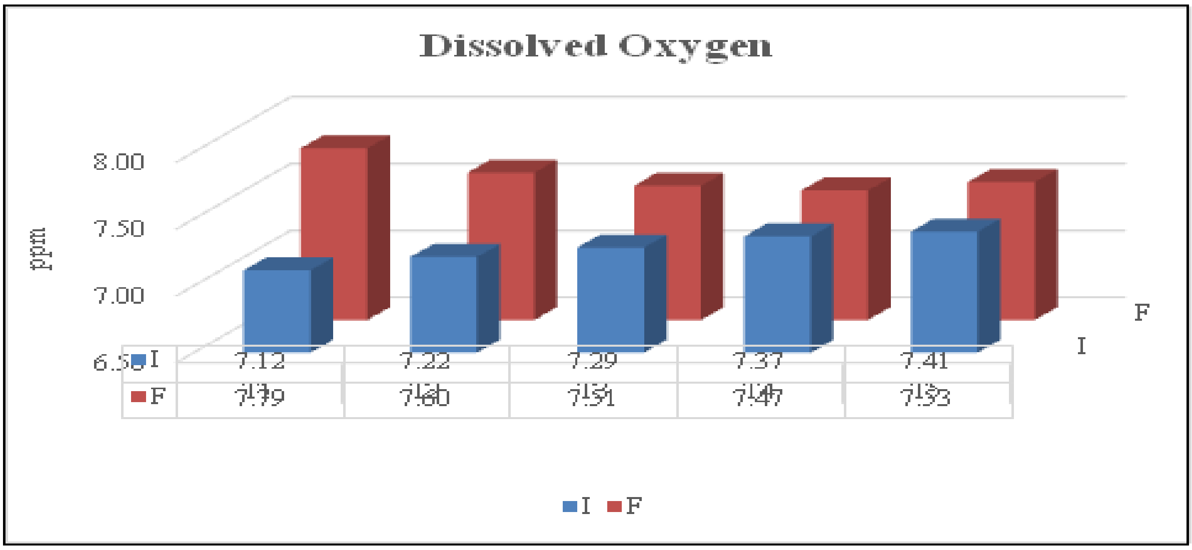

Fig. 12. Dissolved oxygen variation in all the treatments.

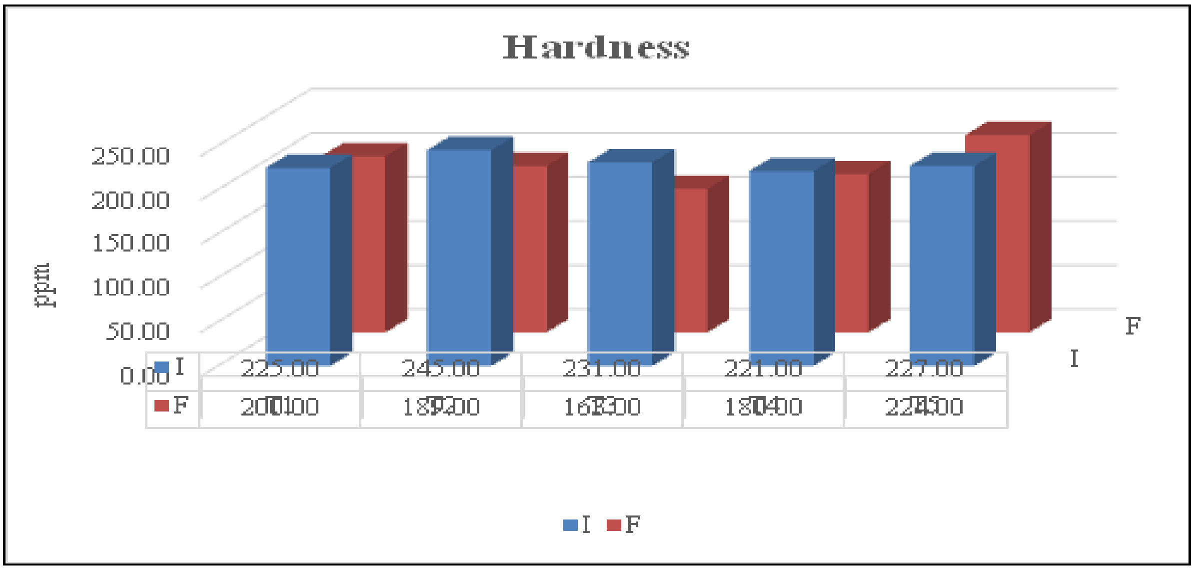

Fig. 13. Hardness variation in all the treatments

Tomohito *et al.*, 2005also investigated the removal of chloride from various solutions including NaCl, HCl, MgCl2, AlCl3, CaCl2, and NH4Cl at 60 °C for 3 h using magnesium–aluminum oxide, Mg0.80Al0.20O1.10, prepared by the thermal decomposition of a hydrotalcite-like compound, $Mg_{0.80}Al_{0.20}(OH)_2(CO_3)_{0.10}\cdot 0.78H_2O$. Abdel-Wahab *et al.*, 2002 described that chloride is a deleterious ionic species in cooling water systems because it promotes corrosion, and most of the scale and corrosion inhibitors are sensitive to chloride concentration in the water. He removed chloride from cooling water by precipitation as calcium chloroaluminate $[Ca_4Al_2Cl_2(OH)_{12}]$ using the ultra-high lime with aluminum (UHLA) process and characterize the equilibrium conditions of calcium chloroaluminate precipitation. Abdel-Wahab *et al.,* 2005 evaluated three alternative aluminum sources i.e. freshly precipitated aluminum hydroxide $Al(OH)_3(s)$, waste alum sludge, and sodium aluminate ($NaAlO_2$) for chloride removal from recycled cooling water using UHLA treatment process. His lab experiments results indicated that at low aluminum doses, chloride removal was similar for all three aluminum sources, however, at high aluminum doses, chloride removal efficiency was higher with $Al(OH)_3$ than with

$NaAlO_2$ or alum sludge at the same lime dose; differences in results could be attributed to differences in pH. Abdel-Wahab *et al.,* 2002 demonstrated a new fundamental equilibrium model for removal of chloride based on a new program called INVRS K integrated with PHREEQC to calculate values of unknown or poorly defined equilibrium or kinetic constants using a Gauss-Newton nonlinear regression routine; the Model predictions indicated that the results could be best described by assuming the formation of a solid solution of calcium chloroaluminate ($Ca_4Al_2Cl_2OH_{12}$), tricalciumhydroxyaluminate (Ca3Al2OH12), and tetracalciumhydroxyaluminate (Ca4Al2OH14). Roya *et al.,* 2013 investigated a method for simultaneous removal of calcium, magnesium and chloride by using Mg0.80Al0.20O1.10 as a Magnesium-Aluminum oxide (Mg–Al oxide) which resulted in simultaneously removal of Cl^-, Mg^{+2} and Ca^{+2} from distiller waste of a sodium carbonate production factory which were characterized with respect to nitrogen physicosorption, X-ray diffraction (XRD) and field emission scan electron microscopy (FESEM) morphology. Tomohito *et al.,* 2003 investigated the removal of Ca^{2+} and Cl^- from $CaCl_2$ solution at 20–60°C, using magnesium–aluminum oxide, Mg0.80Al0.20O1.10. Yingjun *et al.,* 2012 investigated chloride removal of aqueous solution by calcined layered double hydroxides in batch model. Mousa *et al.,* 2012 explained the removal of chloride from the industrial wastewater by adsorption using Amberlite IRA-402 via batch and continuous adsorptions. Orellan *et al.,* 2004 described an electrochemical chloride extraction (ECE) for removal of chloride from a solution of NaCl within 7 weeks. Xuelian *et al.,* 2013 used an electrochemical method to remove chloride ions from zinc sulfate aqueous solution using a potentiostatic technique, and using copper sheets as a working electrode and an auxiliary electrode, Ag/AgCl electrode as a reference electrode.

Conclusion

The chemical method was employed for the removal of chlorides salinity from aquatic media. Four chemicals namely Aluminium hydroxide, Sodium acetate, Aluminium nitrate and Zinc nitrate were used for decreasing chlorides salinity. An increase in the values of all physico-chemical parameters was observed from their initial levels except hardness which was decreased on addition of chemicals.

All the chemicals were found effective in reducing chloride salinity, however, Zinc nitrate was the most efficient in removing chlorides, Aluminium hydroxide was less efficient, then Aluminium nitrate and then Sodium acetate was the least efficient one. For fish survival and growth rate, Aluminium hydroxide was the most effective and Zinc nitrate was the least effective one. Hence, it is recommended to use Aluminium hydroxide for decreasing the aquatic chlorides salinity taken into consideration its better results on fish survival and growth, also.

Acknowledgement

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