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Soil Solution Changes Affected by Biosolids and Aluminum-Based Drinking Water Treatment Residuals (Al-WTRs) During Short-Term Incubation Experiment

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Abstract

An incubation experiment was conducted at Southern Illinois University Edwardsville (SIUE) to investigate the effects of biosolids and WTRs application on soil solution chemistry of Troy soils during incubation time. The results indicated that equilibrium time for each element was different. In general, soil solution pH was significantly increased ($p<0.05$) with increasing WTRs application rates at all incubation times. Similarly, electrical conductivity of soil solution was significantly increased ($p<0.05$) with increasing WTRs application rates at all incubation times. Also, EC of soil solution was significantly increased with increasing WTRs application rates in 50 $g.kg^{-1}$biosolids-amended soils. Large changes in concentrations of different anions with WTRs and /or biosolids application during the incubation time. Calcium concentrations were significantly increased with increasing application rate of WTRs at all incubation The results also indicated that 24 hours may be enough for Cr to equilibrate with the solid phase. The value of Mg concentration at 20 days of incubation was the greatest in biosolids-untreated soils, and then Mg concentrations decreased with increasing incubation time. In biosolids-treated soil, the K concentrations were dramatically increased with increasing incubation time to 20 days at all WTRs application rates. . Na concentrations in biosolids treated and un-treated soils were much higher after 20 days incubation time with a subsequent decrease at 40 and 60 days of incubation. The P concentrations were significantly reduced with increasing WTRs application rates at all incubation times. While, the concentration was significantly increased with increasing the incubation time in both biosolids treated and untreated soils. Cu concentrations in biosolids untreated soils were small and there was a significant difference between incubation times or application rates of WTRs. Also, Ni concentrations in leachate were not detectable in biosolids untreated soils. While, in biosolids-treated soils ,Ni concentrations were significantly increased after 20 days incubation and reached to the maximum value at all treatments of WTRs after 20 days incubation time. In biosolids-treated soils, Mn concentrations in soil solution were much higher than those of un-treated soils. Also, maximum concentrations of Mn were observed after 60 days of incubation time. Mn concentrations were significantly decreased with increasing application rate of WTRs in biosolids and treated and un-treated soils. Zn concentrations were significantly decreased with increasing application rate of WTRs in biosolids treated and un-treated soils. In biosolids untreated soils, Mo, Fe and V were very small concentrations in soil solution during all incubation time. The maximum concentration of Sr was observed after 20 days of incubation, but maximum Ba concentration was recorded after one day of incubation. However, in biosolids-treated soil, Sr concentration reached to the maximum after 20 days and Ba concentration reached to the maximum value after 60 days of incubation. Al concentrations in biosolids untreated soils were very small and there was a significant difference between incubation times or application rates of WTRs. While, in biosolids-treated soils , Al concentrations were significantly increased after 40 days incubation and reached to the maximum value at all treatments of WTRs after 40 days incubation time.

Keywords: Biosolids - composition -Soil solution-WTRs

1. Introduction

Soil solution analysis can be used for prediction of plant response (bioavailability) to nutrients and trace elements in the soil. An understanding of soil solution composition can also be helpful in estimating the speciation and forms of trace elements and nutrients which may be transported into surface and ground water (Sposito, 1984). Trace elements in the soil solution equilibrate with various solid phase components. Therefore soil solution is used to predict binding of nutrients and trace elements to solid phase components.

Trace metal mobility and solubility in soils are of environmental significance due to their potential toxicity to both humans and animals (Chirenje et al., 2003; Ma et al., 1995). Trace metal mobility is closely related to metal solubility, which is further regulated by adsorption, precipitation and ion exchange reactions in soils. Although much effort has been spent on modeling trace metal solubility (Cederberg et al., 1985; Martin et al.,2003; Sposito, 1984), such predictions in field conditions suffer from much uncertainty. This uncertainty is partially due to the difficulty in assessing the effects of dynamic soil solution chemistry on trace metal speciation (Jensen et al., 1999). However, changes in soil solution chemistry, such as pH, redox potential and ionic strength, may also significantly shift the retention processes of trace metals by soils (Gerringa et al., 2001). These effects may be further complicated by ligand competition from other cations (Amrhein et al, 1994; Norrstrom and Jacks, 1998).

Water treatment residuals (WTRs) and biosolids are both by-products from municipal treatment processes. Aluminum-based WTRs/Alum-sludge are considered a waste product from drinking water treatment facilities. Alum $[Al_2(SO_4)_3 \cdot 14H_2O]$ is the main component used in the treatment process for colloid destabilization, flocculation, and water clarification. Biosolids are a by-product of wastewater treatment. Both products have been studied separately for their effects and benefits for land application as an alternative method of beneficial reuse. The benefits of WTR soil application include increased organic C, improved structure, and increased water-holding capacity (Bugbee and Frink, 1985; Elliott et al., 1990; Rengasamy et al., 1980; Mahdy et al., 2009). Land application of biosolids is both environmentally and economically advisable. Biosolids addition provides organic matter (OM) to soil and this addition may represent a good alternative to prevent degradation of soils (Roldan et al., 1996) and to improve many physical properties of agricultural soils such as water holding capacity, aeration, porosity and cation exchange capacity (Engelhart et al., 2000). Moreover, the application of this residue offers the possibility of recycling plant nutrients with the beneficial effects on soil fertility and plant nutrition (Casado-Vela et al. 2007; Gascó and Lobo, 2007).

Knowledge of the chemical composition of the soil solution can be a useful tool for evaluating the effects of biosolids and WTRs amendments. The soil solution is the mobile phase in soils from which plants derive their supply of nutrients and the medium where soil chemical reactions occur (Curtin and Smillie, 1995). There is evidence that the chemistry of the soil solution of biosolids amended soils is a good indicator for solubility, mobility and availability of nutrients and in general for the nutrient status of the soil. Unfortunately, few data on the effects of biosolids and WTRs application on soil solution chemistry are available for neutral and slightly alkaline soils. These data are important to predict nutrient mobility and availability to plants and losses by leaching and may help to elaborate a careful strategy including the use of biosolids and WTRs amendments in management of chemical fertility of slightly alkaline soils. This study was undertaken to investigate the effects of biosolids and WTRs application on the soil solution chemistry of Troy soils using selected incubation times.

2. Materials and Methods

2.1 Description of soil, biosolids, and WTRs

This laboratory incubation experiment was performed with soil collected from an agricultural field in Liberty, Illinois located at 685872E, 44201764N zone 15s. The soil had not been amended with biosolids or animal manures for at least 25 years. Soils were analyzed for general properties according to standard methods (Page et al., 1982) (Table I).

Properties of interest include pH, cation exchange capacity (CEC), OM, P-bicarbonate, and texture.

Biosolids were collected from the Troy Municipal Wastewater Treatment Plant in Troy, Illinois. The water content of the biosolids was 75%. The pH of the biosolids was 11.9±0.2.

Drinking water treatment residuals (WTRs) were collected from the Hartford Drinking Water Treatment Facility in Hartford, Illinois. The WTRs were taken directly from holding tanks, where coagulated particulates and alum and lime treatment are allowed to precipitate from water. Water was drained following collection until the WTRs reached a moisture content of 100%. The pH of the WTRs was 9.0±0.1. WTRs were stored indoors in plastic buckets.

Soils, biosolids, and WTRs were chemically characterized using inductively coupled plasma mass spectroscopy (ICP-MS) to determine concentrations of elements (Poulter-Miller, 2006). Samples were oven-dried at 45ºC for 3 days and ground to a fine powder in an agate mortar and pestle. Ground samples were digested using EPA method 3050B for sludges and soils.

2.2 Sample preparation and experimental design

All samples of soil, biosolids and WTR were ground to a fine powder using a ceramic mortar and pestle. After grinding, each of the three components (biosolids, WTRs, and soil) was weighed appropriately to provide each of the following treatments: WTR rates (0, 20, 40, and 80 g WTRs kg^{-1} soil). Each WTR treatment level was crossed with biosolid loading rates of (0, 25, and 50 g biosolid kg^{-1} soil) (n=3 for each treatment combination). The experimental design was a split -split plot design repeated for each level of biosolids application. This design was chosen because the purpose of the objective of the experiment was to determine the effects of incubation, time, biosolids and WTRs. The treated soils were well mixed with biosolids and WTRs and transferred to a large plastic bin. De-ionized water was added to bring the soil to its field capacity. Then, the treated and control soils were transferred to glass jars. The moisture content of the treated and control soils was kept constant during incubation by calculating the field capacity and periodically weighing the jars and adding de-ionized water to compensate for water loss via evaporation. Jars were covered with perforated plastic covers and incubated at 25 ºC for 60 days. After the incubation period, the soils were air-dried, crushed to pass a 2-mm sieve, and sub- samples were collected for chemical analysis.

2.3 Soil solution extraction

At 1, 20, 40, and 60 days after application of WTRs and/or Biosolids, four 25 g soil were collected and soil solutions were extracted using a rapid centrifugation method (Elkhatib et al., 1987). Soil solutions were filtered through 0.45 µm filters. EC and pH were measured in soil solution at each incubation interval, then analyzed by inductively coupled argon plasma emission spectrometry(ICP-MS) for Ca, Mg, Na, K, P, Fe, Mn, Cu, Zn, Ni, Mo, Cr, Sr, Ba, V, Al and Si and Ion Chromatography(IC) for NO_3,NO_2,PO_4,SO_4,Cl,F,,Br,OH,CO_3,HCO_3.

2.4 Statistical analysis

Statistical analysis was done using analysis of covariance Statistical Analysis Software (SAS, version 8.0) (SAS Institute Inc., 1999). All statistical tests were performed with a significance level of 0.05. Graphs were generated using SigmaPlot (version 10.0) (Systat Software Inc., 2006).

3. Results and Discussion

3.1 Changes in electrical conductivity (EC) and pH with incubation

In general, soil solution pH was significantly increased ($p<0.05$) with increasing WTRs application rates at all incubation times (Table 2).Also, soil pH was significantly increased with increasing WTRs application of 50 $g.kg^{-1}$biosolids-amended soil (Table II).The increase in pH was associated with increased application rates of WTRs and biosolids because the pH of WTRs and biosolids were 9.00 and 11.90 respectively. In general, pH of soil solution was highly changed with change in incubation time (Table II).

For example, in soil treated with 80 $g.kg^{-1}$ WTRs, pH of soil solution increased from 7.94 to 8.02, 8.13 at 20 and 40 days of incubation respectively, then the reduction in pH was recorded. On the contrary, in soil treated with 80 $g.kg^{-1}$ WTRs and 50 $g.kg^{-1}$ biosolids, pH of soil solution increased from 9.78 to 9.81,9.85, and 9.50 at 20,40,and 60 days of incubation(Table II). Similarly, In general, electrical conductivity of soil solution was significantly increased ($p<0.05$) with increasing WTRs application rates at all incubation times (Table II).Also, EC of soil solution was significantly increased with increasing WTRs application rates in 50 $g.kg^{-1}$biosolids-amended soil (Table II). In general, EC of soil solution was highly changed with change in incubation time(Table II).For example, in soil treated with 80 $g.kg^{-1}$ WTRs, EC of soil solution increased from 846.50 to 866.53,880.12, and 895.15 μScm^{-1} at 20 ,40 and 60 days of incubation respectively. On the contrary, in soil treated with 80 $g.kg^{-1}$ WTRs and 50 $g.kg^{-1}$ biosolids, EC of soil solution increased from 3830 to 4000,4100, and 4185 μScm^{-1} at 20,40,and 60 days of incubation(Table II). It was obvious that WTRs and biosolids application affects the soil solution chemistry in two ways, as a liming agent and as a supplier of nutrients. As a liming agent, WTRs and biosolids application induced increases in soil solution pH as suggested by many authors in the case of lime application (Bakker et al., 1999; Derome and Saarsalmi, 1999; Hildebrand and Schack-Kirchner, 2000). As a supplier of elements, the increase in the soil solution pH with WTRs and biosolids was partly due to ligand exchange between WTRs and biosolids SO4 and OH ions (Alva and Sumner, 1990).

3.2 Changes in soil solution composition during incubation

Soil solution composition reflects the intensity and distribution of trace elements in the soil aqueous phase and represents the integration of multiple physical, chemical, and biological processes occurring concurrently within the soil (Sposito, 1984).However, composition reflects the soil moisture content, sample handling, and the displacement technique. Major cations and anions in soil solution include Ca,Mg,K,Na,NO_3,Cl,SO_4,HCO_3,CO_3, and H2PO_4, of which Ca and Mg are the dominant soil solution cations and HCO_3 and CO_3 are the dominant anionic species(Hirsh and Banin,1990).

Large changes in concentrations of different anions with WTRs and /or biosolids application during the incubation time were noticed(Table II).NO_3 concentrations were significantly increased with increasing application rate of WTRs at all incubation times. The value of NO_3 concentration at 80 gkg^{-1} was 314.33 mgl^{-1} after one day incubation, then increased to361 mgl^{-1} at 20 days of incubation and the maximum value was recorded at 60 days of incubation .Similarly, in biosolids-amended soil the NO_3 concentrations were much higher than those in untreated biosolids soil (Table II).In biosolids-treated soil, the nitrate concentrations were dramatically increased with increasing WTRs application rates. For example, the nitrate concentrations increased from 124.67 to 378.67 mgl^{-1} after one day of incubation when 80 $g.kg^{-1}$ WTRs was applied, and reached to 394.67 after 20 days of incubation. But, the value was 408.33 and 427 mgl^{-1} after 40 and 60 days of incubation at the same application rate of WTRs in 50 $g.kg^{-1}$ biosolids-treated soil (Table II). The nitrite concentrations were below 0.01 mgl^{-1} at all application rates of WTRs and biosolids at all times of incubation (Table II). The biosolids treatment, WTRs rates, incubation time and their interaction significantly affected nitrate concentrations in soil solution (Table II). The relative higher

NO_3 levels in soil solution from WTRs and/or biosolids amended soils suggest that co-application of WTRs and biosolids enhanced nitrification more efficiently than no co-application. The improvement of nitrification seemed to be the result of induced output of DOC since, according to Hildebrand and Schack-Kirchner (2000), this output is a potential energy source for heterotrophic nitrifiers.

In contrast, the orthophosphate concentrations were significantly reduced with increasing WTRs application rates at all time of incubation (Table II).While, the concentration was significantly increased with increasing the incubation time. For example, in biosolids-treated soil, the concentration of phosphate increased from 0.6 to 0.67 and 0.85 at 1, 20, and 40 days of incubation respectively at 80 $g.kg^{-1}$ of WTRs. However, in biosolids-untreated soil, the phosphate concentrations were much lower than those of biosolids-treated soil (Table II). The biosolids treatment, WTRs rates, incubation time and their interaction significantly affected phosphate concentrations in soil solution (Table II).

Many researchers have been investigating the ability of WTRs to reduce the mobility and promote retention of P in impacted soils (Peters and Basta, 1996; O'Connor et al., 2002; Dayton et al., 2003; Ippolito et al., 2003; Makris et al., 2004; Novak and Watts, 2004; Dayton and Basta, 2005; Elliot et al., 2005; Makris et al., 2005; Novak and Watts, 2005a; Silveria et al., 2006; Agyin-Birikorang et al., 2007, Agyin-Birikorang and O'Connor, 2007; Mahdy et al., 2007,Huff,2010). It has been demonstrated that WTR can promote P retention when applied to biosolids-amended soils. However, there are no field studies to date that focus specifically on co-application of biosolids and WTRs to silt loam agricultural soils. Ippolito et al. (1999) and Bayley et al. (2008) reported that WTRs can improve P retention in field conditions but previous studies have focused primarily on sandy or semi-arid soils.

In contrast of phosphate, sulphate anion concentrations were dramatically increased during incubation time and with increasing application rate of WTRs in biosolids treated and untreated soils (Table II).In biosolids untreated soil, SO_4 concentration increased from 13.23 to 45.73 $mg.l^{-1}$ after one day of incubation when the WTRs applied with 80 $g.kg^{-1}$.While, the concentration increased to 83,126.67, and 273.67 $mgt.l^{-1}$ after 20, 40, and 60 days of incubation respectively. Similarly, in biosolids-treated soil, SO_4 concentrations in soil solution increased from 65 to 243.67, from 86.67 to 292.67, from 170.75 to 338.33, and from 273.67 to 546 $mg.l^{-1}$ at 1,20,40,and 60 days of incubation respectively(Table II). The biosolids treatment, WTRs rates, incubation time and their interaction significantly affected SO_4 concentrations in soil solution (Table II). The increase in SO_4 concentrations in soil solution occurred with WTRs application was due to the high concentration of aluminum sulphate in WTRs.

In general, Cl and F concentrations in soil solution were significantly decreased with increasing WTRs application rates in biosolids treated and untreated soils at all incubation times(Table II).However, the concentrations of them were significantly increased with increasing of incubation time. The maximum value for chloride concentration was noticed at 80 $g.kg^{-1}$ WTRs in 50 $g.kg^{-1}$ biosolids-treated soil after 60 days of incubation. Similarly, the maximum value for fluoride concentration was noticed at 80 $g.kg^{-1}$ WTRs in 50 $g.kg^{-1}$ biosolids-treated soil after 60 days of incubation. The biosolids treatment, WTRs rates, incubation time and their interaction significantly affected Cl and F concentrations in soil solution (Table II).

Br, OH, and CO_3 concentrations in soil solution were measured, but concentration was below 0.01, 0.10, and 0.10 $mg.l^{-1}$ for Br, OH, and CO_3 respectively at all treatments and incubation times (Table II).

Finally, HCO_3 concentrations in soil solution were dramatically increased with increasing biosolids rate, WTRs rates, and incubation time (Table II).For instant, in biosolids untreated soil, HCO_3 concentration increased from 62.60 to 480 $mg.l^{-1}$ after one day of incubation when the WTRs applied with 80 $g.kg^{-1}$.While, the concentration increased to 517,620.33, and 644.33 $mgt.l^{-1}$ after 20, 40, and 60 days of incubation respectively. Similarly, in biosolids-treated soil, HCO_3 concentrations in soil solution increased from 807.33 to 984.67, from 925.67 to 1092.33, from 962.25 to 1149, and from 1025 to 1473.33 $mg.l^{-1}$ at 1,20,40,and 60 days of incubation respectively(Table II). The biosolids treatment, WTRs rates, incubation time and their interaction significantly affected HCO_3 concentrations in soil solution (Table II). The increase in HCO_3 concentrations in soil solution that occurred with WTRs and biosolids application was due to the high concentration of lime in WTRs. These results were coincide with the results of Hirsh and

Banin,(1990) who reported that HCO_3 and CO_3 are the dominant anionic species in soil solution of semi-arid soil (Hirsh and Banin,1990).In atypical arid soil with pH 8.0 (California, US), Villarroel et al,(1993) found that Cl>SO_4>NO_3>HCO_3 and Cl in soil solution of salt soils ranges from a few milligrams per liter to several hundred milligram per liter.

Also, large changes in concentrations of elements with WTRs and /or biosolids application during the incubation time were noticed(figs 1-3).Ca concentrations were significantly increased with increasing application rate of WTRs at all incubation times. The value of Ca concentration at 20 days of incubation was the greatest in biosolids-untreated soils, then Ca concentrations were decreased with increasing of incubation time (Fig.1 A).In biosolids-amended soil the Ca concentrations were much higher than those in untreated biosolids soil (Fig.1 A).In biosolids-treated soil, the calcium concentrations were dramatically increased with increasing incubation time to 20 days at all WTRs application rates. The maximum value of Ca was recorded (about 580 $mg.l^{-1}$) at soils treated with 50 $g.kg^{-1}$ biosolids and 20 $g.kg^{-1}$ WTRs. The relative higher Ca levels in soil solution from WTRs and/or biosolids amended soils suggest that co-application of WTRs and biosolids enhanced solubility of lime and release more soluble calcium because WTRs and biosolids contain more lime which was added during treatment of both.

In contrast, Mg concentrations in biosolids untreated soils were much higher than those of biosolids-treated soils(Fig.1 B).In general, Mg concentrations were significantly increased with increasing application rate of WTRs at all incubation times. The value of Mg concentration at 20 days of incubation was the greatest in biosolids-untreated soils, then Mg concentrations were decreased with increasing of incubation time (Fig.1 B).However, in biosolids-amended soil, Mg concentrations were much higher after 20 days incubation time and reached to maximum value after 60 days of incubation (Fig.1 B). The results clarified that 60 days of incubation were the best period for Mg equilibrium in soils treated with biosolids and WTRs. Similar results were reported in different studies. Villarroel et al.,(1993) studied a typical arid soil with pH 8.0(California, USA) and reported that Ca and Mg concentrations in soil saturation extracts range from 1.0-2.0 x 10^{-3} M, and Ca concentrations was higher than Mg concentrations in soil extracts.

Similarly to Ca, Large changes in concentrations of K with WTRs and /or biosolids application during the incubation time. K concentrations were significantly increased with increasing application rate of WTRs at all incubation times. The value of K concentration at 20 days of incubation was the greatest in biosolids-untreated and treated soils, then K concentrations were decreased with increasing of incubation time (Fig.1 C).In biosolids-amended soil, K concentrations were much higher than those in untreated biosolids soil (Fig.1 C).In biosolids-treated soil, the K concentrations were dramatically increased with increasing incubation time to 20 days at all WTRs application rates. The maximum value of K was recorded (about 50 $mg.l^{-1}$) at soils treated with 50 $g.kg^{-1}$ biosolids and no WTRs.

Also, Na concentrations in biosolids amended soils were higher than those of biosolids un-amended soils(Fig.1 D).In general, increasing of WTRs application rates to 20 $g.kg^{-1}$ significantly increased Na concentrations at all incubation times, but 40 $g.kg^{-1}$ application rate of WTRs reduced Na concentrations. Na concentrations in biosolids treated and un-treated soils were much higher after 20 days incubation time, then much reduction in Na concentrations were noticed at 40 and 60 days of incubation (Fig.1 D)

In contrast, the P concentrations were significantly reduced with increasing WTRs application rates at all time of incubation(Fig.1 E).While, the concentration was significantly increased with increasing the incubation time in both biosolids treated and untreated soils. For example, in biosolids-treated soil, the concentration of phosphate increased after 40 days of incubation respectively at 80 $g.kg^{-1}$ of WTRs. However, in biosolids-untreated soil, the phosphate concentrations were much lower than those of biosolids-treated soil (Fig.1 E).

Many researchers have been investigating the ability of WTRs to reduce the mobility and promote retention of P in impacted soils (Peters and Basta, 1996; O'Connor et al., 2002; Dayton et al., 2003; Ippolito et al., 2003; Makris et al., 2004; Novak and Watts, 2004; Dayton and Basta, 2005; Elliot et al., 2005; Makris et al., 2005; Novak and Watts, 2005a; Silveria et al., 2006; Agyin-Birikorang et al., 2007, Agyin-Birikorang and O'Connor, 2007; Mahdy et al., 2007,Huff,2010).

In biosolids untreated soils, Cr was not detectable during all incubation time(Fig.2 A).While, in biosolids-treated soils ,Cr concentrations were significantly increased after one day incubation and reached to the maximum value at all treatments of WTRs after one day incubation time(Fig.2 A). Cr concentrations were reduced with increasing of incubation time to 20 days, and then increased at 40 and 60 days of incubation but, still lower than that after one day incubation. The results indicated that 24 hours may be enough for Cr to equilibrate with solid phase.

Similarly, Cu concentrations in biosolids untreated soils were very small and there was a significant difference between incubation times or application rates of WTRs (Fig.2 B). While, in biosolids-treated soils, Cu concentrations were significantly increased after one day incubation and reached to the maximum value at all treatments of WTRs after one day incubation time (Fig.2 B). Cu concentrations were reduced with increasing of incubation time to 20 days, and then increased at 40 and 60 days of incubation but, still lower than that after one day incubation. These results on soil solution chemistry were reported. Fotoval et al.,(1997) reported that Zn and Cu concentrations in soil solution of soils from South Australia with pH 7.59-8.99 range from 0.009-0.218 and 0.058-4.43 µM/kg respectively. In German non-contaminated soil with 2.3 % CaCO3 and soil pH 8.5, Zn, Cu, and Cd are 1.87, 0.66, and 0.20 µM/L in soil solution, respectively (Helal et al., 1996).

Also, Ni concentrations in leachate were not detectable in biosolids untreated soils (Fig.2 C). While, in biosolids-treated soils, Ni concentrations were significantly increased after 20 days incubation and reached to the maximum value at all treatments of WTRs after 20 days incubation time (Fig.2 C). Ni concentrations were reduced with increasing of incubation time to 40 and 60 days of incubation. There was no difference in Ni concentrations between 40 and 60 days after incubation.

On the contrary, Mn concentrations were significantly increased during incubation time at all treatments of WTRs in biosolids un-treated soil (Fig.2 D).In general, the maximum concentration of Mn was observed after 60 days of incubation. In biosolids-treated soils, Mn concentrations in soil solution were much higher than those of biosolids un-treated soils. Also, maximum concentrations of Mn were observed after 60 days of incubation time. Mn concentrations were significantly decreased with increasing application rate of WTRs in biosolids and treated and un-treated soils (Fig.2 D).

In general, Zn concentrations in soil solution were significantly increased with increasing time of incubation to 20 days (Fig.2 E).However, more reduction in Zn concentrations was found at 40 and 60 days of incubation in biosolids treated or un-treated soils. Zn concentrations were significantly decreased with increasing application rate of WTRs in biosolids and treated and un-treated soils (Fig.2 E).

Si concentrations in soil solution were significantly increased with increasing of incubation time (Fig.2 F).Application of WTRs with different rates increased Si concentrations in biosolids treated and untreated soils. The maximum value of Si concentrations was recorded at 50 $g.kg^{-1}$ biosolids-treated soil treated with 20 $g.kg^{-1}$ WTRs (Fig.2 F). The results indicated that equilibrium between soil solution and solid phase for Si was 60 days.

In biosolids untreated soils, Mo, Fe and V were small concentrations in soil solution during all incubation time (Fig.3 A, B, and C). While, in biosolids-treated soils, Mo, Fe and V concentrations were significantly increased after 20 days, 40 days and one day incubation time for Mo, Fe and V, respectively. In general, increasing of WTRs application rates reduced concentrations of Mo, Fe and V in soil solution. The significant reduction of aqueous Fe concentrations during incubation was probably a result of FeCO3 precipitation. On the other hand Sr and Ba concentrations were detectable in biosolids untreated soil (Fig.3 D and E). The maximum concentration of Sr was observed after 20 days of incubation, but maximum Ba concentration was recorded after one day of incubation. However, in biosolids-treated soil, Sr concentration reached to the maximum after 20 days and Ba concentration reached to the maximum value after 60 days of incubation. Addition of WTRs with different rates increased Sr and Ba concentrations in soil leachate. Similarly, Al concentrations in biosolids untreated soils were very small and there was a significant difference between incubation times or application rates of WTRs (Fig.3 F). While, in biosolids-treated soils, Al concentrations were significantly increased after 40 days incubation and reached to the maximum value at all treatments of WTRs after 40 days incubation time (Fig.3 F). Al concentrations were increased with increasing of WTRs application rate up 20 $g.kg^{-1}$, then reduced at 40 and 80 $g.kg^{-1}$ application rate.

This results were disagree with other results which reported that Al concentrations were significantly increased with increasing of WTRs application rates(Mahdy et al.,2009).

Effects of WTRs on trace elements availability in biosolids-treated soils has been studied by many researchers. Makris et al.,(2009) reported that the Al-WTRs was highly effective in removing both As(v) and As(III). The study of Hovsepyan et al.,(2009) indicated that a strong affinity of Hg for Al-WTRs and can be used to remove Hg from aqueous solutions. This ability points to the potential of Al-WTRs as a sorbent in soil remediation techniques based on Hg-immobilization. Nagar et al.,(2010) have been proposed the Al-WTRs as a low-cost alternative sorbent for arsenic (As)-contaminated aquatic and soil system and they found that the Al-WTRs demonstrated 100% As(V) sorption in the entire pH range. Brown et al.,(2005) found that both soil solution and ammonium nitrate extractable heavy metal(Cd, Pb, and Zn) were decreased by all treatments included lime, P, red mud, cyclonic ashes, biosolids, and water treatment residuals. In a similar experiment, Amrhein et al. (1994) used soils incubated under water-flooded conditions, and found increased Fe (II) concentrations in pore water with incubation time possibly as a result of reductive Fe dissolution (Amrhein et al., 1994). Finally, it can be concluded that the reduction in soil solution trace elements concentrations resulting from the application of WTRs to biosolid-amended soils can be explained by formation of metal-sulfate, low solubility product, and the floc-adsorption and the co-precipitation processes, in which the formation of a mixed solid phase by the incorporation of metal ions into the crystal lattice of another precipitating solid phase is expected (Karthikyan et al., 1996).

4. Conclusion

Trace metal mobility and solubility in soils are of environmental significance due to their potential toxicity to both humans and animals (Chirenje et al., 2003; Ma et al., 1995). Trace metal mobility is closely related to metal solubility, which is further regulated by adsorption, precipitation and ion exchange reactions in soils. Numerous researchers have reported on changes in solubility and mobility of trace metals with incubation. However, reported data have often been contradictory. This is in part due to the differences in soil/water ratios used in various experiments including metal concentrations determined in pore water, in filtrates separated from soil suspensions, and in leachate from a soil leached with pure water or an electrolyte. Additionally, changes in aqueous trace metal concentrations with incubation reported in literature can be confusing. For example, metal concentrations in pore water or metal mobility leached by pure water may decrease with incubation when no precipitation is observed. Therefore, the roles of Fe in controlling metal concentrations in these systems need to be considered. We suggested that CEC of a soil may change with incubation underwater-flooding condition, and thus might greatly affect metal solubility and mobility.

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Table 1. General properties of the experimental biosolids, WTRs, and soils. Values are means ± standard deviation (n=3).

Element	Units	WTRs	Biosolids	Soil
pH		9.00±0.05	11.9±0.2	6.27±0.12
EC	dSm^{-1}	1.88±0.05	6.15±0.11	1.05±0.07
Texture		nd	nd	Silty loam
O.M	$g\ kg^{-1}$	67.6±0.5	380.2±3.2	20.3±0.6
Available-N	$mg\ kg^{-1}$	19.06±1.56	78.09±1.05	23.34±0.59
KCl-Al	$mg\ kg^{-1}$	125.07±4.5	34.67±0.58	75.23±1.52
Available-P	$mg\ kg^{-1}$	17.12±0.54	53.99±2.51	26.00±4.00
CEC	$cmol(+)kg^{-1}$	43.12±5.54	76.00±3.5	10.53±0.06
ICP-MS analysis:				
Aluminum (Al)	$g\ kg^{-1}$	12.6±0.05	71.9±0.7	71.0±0.47
Sodium (Na)	$g\ kg^{-1}$	0.5±0.02	1.5±0.02	<0.001
Iron (Fe)	$g\ kg^{-1}$	121.0±4.0	82.3±2.9	123.2±4.9
Potassium (K)	$g\ kg^{-1}$	0.5±0.01	30.7±0.8	10.6±0.6
Magnesium (Mg)	$g\ kg^{-1}$	12.6±0.5	3.6±0.3	1.5±0.02
Silver (Ag)	$mg\ kg^{-1}$	<0.002	2.00±0.12	<0.002
Arsenic (As)	$mg\ kg^{-1}$	<0.1	11.51±0.26	3.80±0.19
Boron (B)	$mg\ kg^{-1}$	12.30±0.58	109.07±5.06	6.55±1.05
Calcium (Ca)	$mg\ kg^{-1}$	266.07±2.28	309.34±11.28	1.61±0.06
Cadmium (Cd)	$mg\ kg^{-1}$	0.05±0.00	1.57±0.06	0.37±0.03
Cobalt (Co)	$mg\ kg^{-1}$	0.44±0.04	3.00±0.02	7.53±0.37
Chromium (Cr)	$mg\ kg^{-1}$	3.83±0.08	23.72±0.69	12.82±0.59
Copper (Cu)	$mg\ kg^{-1}$	0.86±0.10	342.07±6.70	11.34±0.60
Manganese (Mn)	$mg\ kg^{-1}$	7021.85±279.71	3321.74±63.71	7822.82±444.63
Molybdenum (Mo)	$mg\ kg^{-1}$	0.03±0.04	4.60±0.18	0.45±0.07
Nickel (Ni)	$mg\ kg^{-1}$	7.75±0.71	30.15±0.59	13.50±0.55
Lead (Pb)	$mg\ kg^{-1}$	0.05±0.00	29.16±0.32	20.30±0.54
Zinc (Zn)	$mg\ kg^{-1}$	10.95±0.85	190.86±0.83	64.19±2.15

Table 2. EC, pH, and anion concentrations in soil solution of Troy soil amended with WTRs and Biosolids[a].

Biosolids, $g.kg^{-1}$	WTRs, $g.kg^{-1}$	EC,	pH	NO_3	NO_2	P	SO_4	Cl	F	Br	OH	CO_3	HCO_3
		μScm^{-1}	mg.l^{-1}										
		1-day Incubation											
0	0	846.50±13.02	6.39±0.06	68.50±1.32	<0.01	0.15±0.03	13.23±0.75	18.20±0.72	0.82±0.07	<0.01	<0.10	<0.10	62.60±1.35
	20	950.86±18.09	7.61±0.04	255.67±4.04	<0.01	0.11±0.01	25.70±2.04	14.47±0.61	0.64±0.14	<0.01	<0.10	<0.10	366.67±4.16
	40	1245.00±15.00	7.87±0.04	305.67±5.13	<0.01	0.09±0.01	31.37±2.63	14.27±1.10	0.40±0.10	<0.01	<0.10	<0.10	390.00±5.00
	80	1347.00±14.08	7.94±0.06	314.33±6.03	<0.01	0.05±0.01	45.73±1.55	8.57±0.50	0.13±0.06	<0.01	<0.10	<0.10	480.00±4.58
50	0	3830.00±31.07	9.36±0.05	124.67±5.03	<0.01	1.64±0.08	65.00±4.00	42.50±2.23	34.23±0.84	<0.10	<0.10	<0.10	807.33±7.37
	20	3910.00±89.09	9.46±0.02	277.00±11.00	<0.01	1.20±0.10	129.00±3.00	35.67±1.53	25.67±2.52	<0.10	<0.10	<0.10	884.00±52.46
	40	3990.00±40.05	9.56±0.04	331.67±7.64	<0.01	0.83±0.03	164.00±4.58	28.00±1.00	17.33±1.53	<0.10	<0.10	<0.10	910.00±10.00
	80	4100.00±100.05	9.78±0.02	378.67±8.08	<0.01	0.60±0.03	243.67±7.23	22.33±2.52	16.33±1.53	<0.10	<0.10	<0.10	984.67±14.05
Biosolids	**WTRs**	**20-days Incubation**											
0	0	866.53±33.11	7.56±0.02	93.00±2.65	<0.01	0.23±0.03	35.17±2.25	41.80±3.30	1.20±0.10	<0.10	<0.10	<0.10	82.33±2.08
	20	980.11±19.88	7.81±0.03	291.67±3.79	<0.01	0.18±0.01	48.37±2.97	35.93±3.36	0.84±0.05	<0.10	<0.10	<0.10	394.00±5.29
	40	1280.00±20.00	7.92±0.03	320.67±9.02	<0.01	0.12±0.02	56.97±2.70	29.07±0.93	0.64±0.02	<0.10	<0.10	<0.10	442.00±6.24
	80	1375.00±45.00	8.02±0.04	361.33±7.77	<0.01	0.09±0.01	83.00±5.57	23.00±1.00	0.26±0.05	<0.10	<0.10	<0.10	517.00±6.56
50	0	4000.00±80.00	9.47±0.03	145.00±6.00	<0.01	1.85±0.02	86.67±2.52	82.27±2.82	43.67±4.51	<0.10	<0.10	<0.10	925.67±7.37
	20	4085.00±55.00	9.52±0.03	305.33±5.03	<0.01	1.45±0.17	160.67±5.03	78.10±1.91	33.00±3.00	<0.10	<0.10	<0.10	984.00±10.58
	40	4175.00±85.00	9.60±0.15	344.67±5.03	<0.01	1.18±0.04	203.00±2.65	60.33±2.52	21.67±2.52	<0.10	<0.10	<0.10	1004.00±4.58
	80	4250.00±50.00	9.81±0.03	394.67±5.13	<0.01	0.67±0.04	292.67±3.51	54.00±1.00	16.67±1.53	<0.10	<0.10	<0.10	1092.33±6.66
Biosolids	**WTRs**	**40-days Incubation**											
0	0	880.12±9.88	7.36±0.04	162.67±9.29	<0.01	0.35±0.03	63.67±2.08	71.33±3.21	2.27±0.25	<0.10	<0.10	<0.10	268.00±9.54
	20	995.00±10.00	7.75±0.04	296.33±3.79	<0.01	0.24±0.02	85.33±4.73	60.67±2.08	1.48±0.06	<0.10	<0.10	<0.10	465.33±11.02
	40	1320.00±60.00	8.19±0.02	360.67±5.51	<0.01	0.15±0.03	102.33±3.06	56.67±1.53	0.88±0.03	<0.10	<0.10	<0.10	586.33±5.51
	80	1398.00±47.00	8.13±0.03	392.67±4.73	<0.01	0.08±0.01	126.67±3.51	51.43±3.61	0.36±0.05	<0.10	<0.10	<0.10	620.33±9.29
50	0	4100.00±80.00	9.53±0.03	159.00±2.52	<0.01	2.20±0.12	170.75±5.29	137.50±9.87	49.00±3.06	<0.10	<0.10	<0.10	962.25±5.51
	20	4180.00±50.00	9.57±0.03	325.33±4.16	<0.01	1.46±0.09	204.33±5.13	121.00±4.36	48.33±1.53	<0.10	<0.10	<0.10	1009.00±6.56
	40	4290.00±50.00	9.64±0.02	355.00±11.00	<0.01	1.25±0.08	244.33±6.03	104.33±4.04	26.00±2.65	<0.10	<0.10	<0.10	1085.33±5.03
	80	4380.00±70.00	9.85±0.03	408.33±3.51	<0.01	0.85±0.06	338.33±7.64	84.33±5.51	17.67±1.53	<0.10	<0.10	<0.10	1149.00±6.24
Biosolids	**WTRs**	**60-days Incubation**											
0	0	895.15±84.85	7.12±0.03	192.33±3.06	<0.01	0.45±0.04	120.67±3.06	79.67±2.52	2.86±0.09	<0.10	<0.10	<0.10	298.00±2.65
	20	1010.18±39.72	7.85±0.03	306.00±3.61	<0.01	0.30±0.03	172.67±4.51	64.67±1.53	1.88±0.02	<0.10	<0.10	<0.10	491.33±6.66
	40	1390.00±40.00	7.91±0.02	389.33±5.13	<0.01	0.11±0.01	216.00±5.57	57.33±2.08	1.28±0.05	<0.10	<0.10	<0.10	606.00±6.00
	80	1430.00±25.00	7.95±0.03	432.67±19.66	<0.01	0.06±0.02	273.67±5.13	53.33±1.53	0.74±0.07	<0.10	<0.10	<0.10	644.33±6.03
50	0	4185.00±45.00	9.40±0.09	182.33±6.66	<0.01	2.45±0.06	378.00±14.53	175.33±5.69	57.00±2.65	<0.10	<0.10	<0.10	1025.00±4.36
	20	4260.00±40.00	9.42±0.04	346.50±3.54	<0.01	1.25±0.13	440.50±6.36	147.50±3.54	50.50±0.71	<0.10	<0.10	<0.10	1322.00±2.83
	40	4400.00±60.00	9.46±0.05	387.67±8.74	<0.01	0.81±0.09	483.33±5.69	125.33±2.08	37.33±1.53	<0.10	<0.10	<0.10	1389.00±10.15
	80	4600.00±50.00	9.50±0.04	427.00±3.61	<0.01	0.36±0.04	546.00±3.61	109.67±7.37	23.67±1.53	<0.10	<0.10	<0.10	1473.33±30.17
$LSD_{0.05}$(WTRs)		**41.98**	**0.03**	**3.82**	-	**0.05**	**2.88**	**2.09**	**0.98**	-	-	-	**6.01**
$LSD_{0.05}$(biosolids)		**29.73**	**0.01**	**4.87**	-	**0.01**	**1.95**	**2.28**	**1.31**	-	-	-	**12.75**
$LSD_{0.05}$(incubation)		**26.00**	**0.02**	**4.07**	-	**0.04**	**3.02**	**1.90**	**0.90**	-	-	-	**7.43**
ANOVA		***F-test***											
Biosolids		***[b]	***	**	-	***	***	***	***	-	-	-	***
WTRs		***	***	***	-	***	***	***	***	-	-	-	***
Incubation time		***	***	***	-	***	***	***	***	-	-	-	***
WTRs x Biosolids		***	***	**	-	***	***	***	***	-	-	-	***
WTRs x Incubation		***	***	***	-	***	***	***	***	-	-	-	***
Biosolids x Incubation		***	NS[c]	***	-	***	***	***	***	-	-	-	***
WTRs x Biosolids x Incubation		***	NS	***	-	***	***	***	***	-	-	-	***

[a] Data are the average (n=3) ± standard deviation .
[b] *** are significant at 0.001 probability level.
[c] NS: non significant

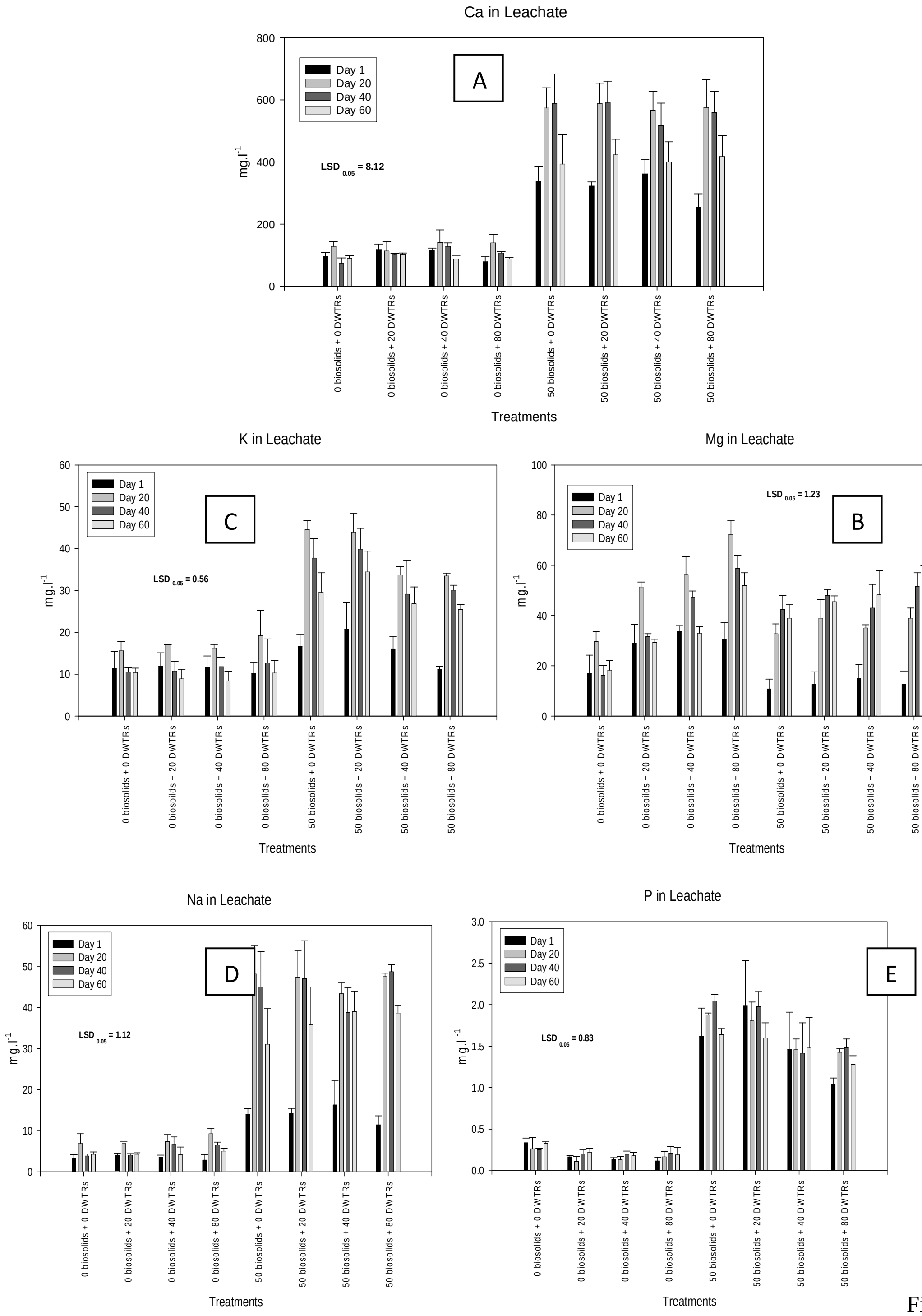

Fig.(1):Ca, Mg, K, Na, and P concentrations in soil solution of Troy soil amended with WTRs and Biosolids. Where no error bars are present, the standard error was too small to be represented as the scale of the diagram

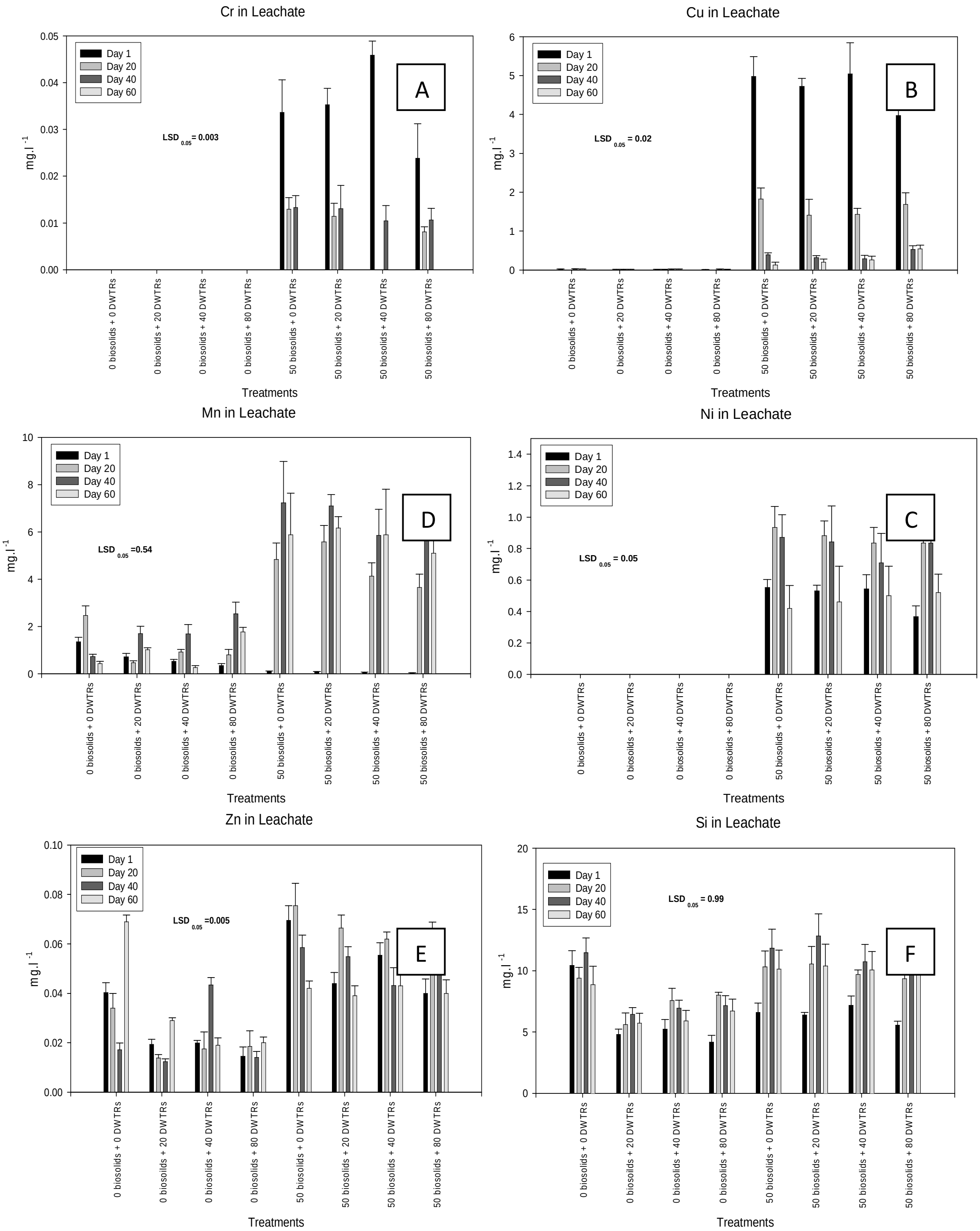

Fig.(2): Cr, Cu, Ni, Mn, Zn, and Si concentrations in soil solution of Troy soil amended with WTRs and Biosolids. Where no error bars are present, the standard error was too small to be represented as the scale of the diagram

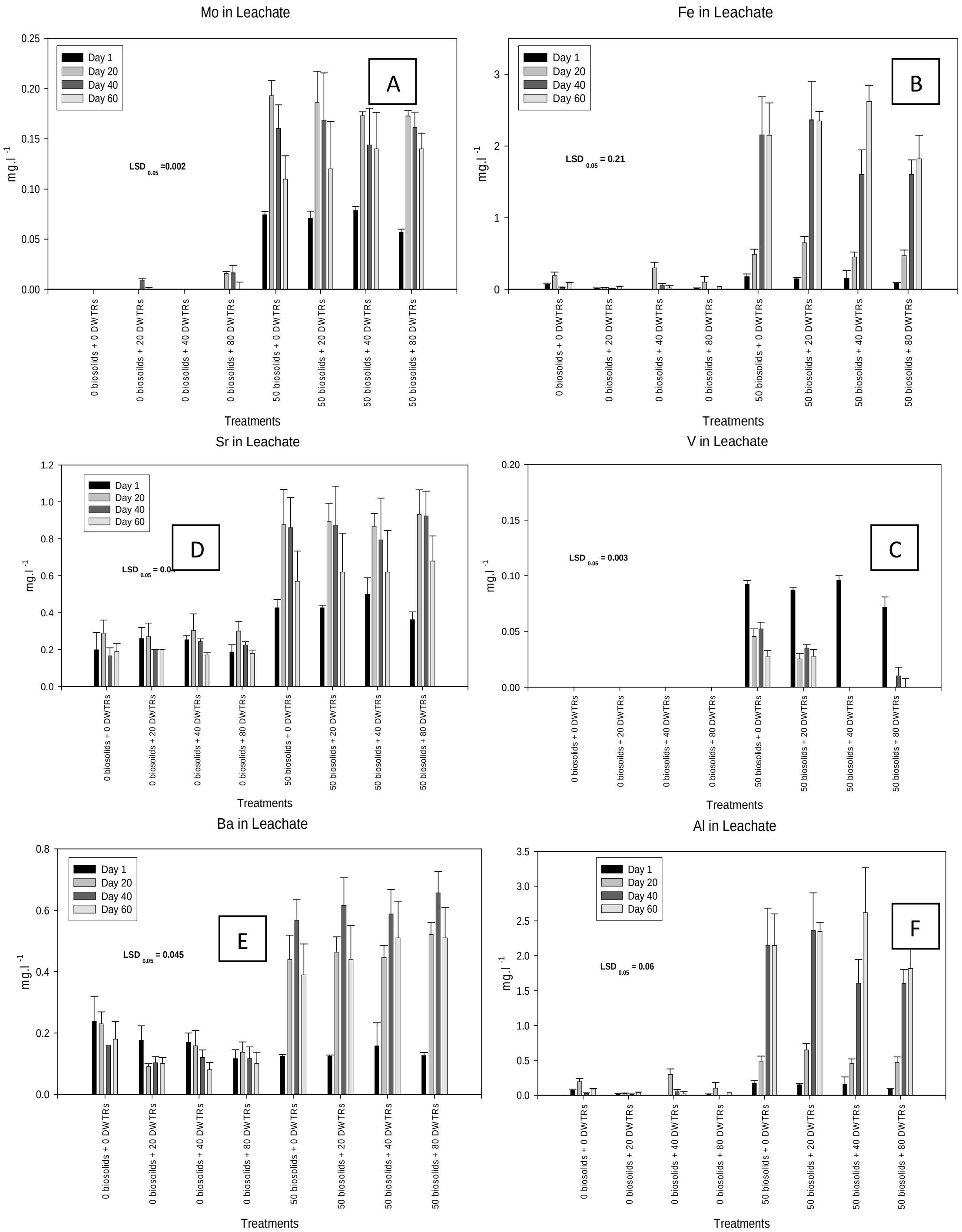

Fig.(3):): Mo, Fe, V, Sr, Ba, and Al concentrations in soil solution of Troy soil amended with WTRs and Biosolids. Where no error bars are present, the standard error was too small to be represented as the scale of the diagram