Investigation of Heavy Metal Pollution Caused By Fertilizer in Groundwater with a New Approach: Göksu Delta, Turkey

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ABSTRACT

Today the chemical fertilizers used in agricultural applications have been an important factor pollution of environmental. Göksu Delta is an important wetland where the Göksu River reaches to sea in the eastern of the town Taşucu-Mersin. The delta is classified as a Wetland of International Importance according to the Ramsar Convention. In this area, use of excessive fertilizer effects the surface and groundwater quality negative. Because of the Delta's geological structure, soil characteristics and climatic conditions, groundwater quality is affected very easy from the surface activities. In the agricultural activities intensively used fertilizers and pesticides contain not only N and P compounds but also some heavy metals. This study aim is investigation of heavy metal pollution sourced from fertilizers in groundwater with a new approach. For this, the most widely used fertilizer in the area (ammonium nitrate) and a groundwater samples were taken at three different time periods and were analyzed (electrical conductivity (EC), salinity (Sal), temperature(T), pH, iron (Fe), copper (Cu), chromium (Cr), manganese (Mn), nickel (Ni), zinc (Zn), cadmium (Cd), lead (Pb)). The results were transferred to Scholler semi-logarithmic diagram which is a new approach in identifying pollutant sources to show the effect of fertilizer. In addition to, the results were compared with the limit values in order to determinate the quality of groundwater. Especially, samples that were taken in the rainy season (1.Period), with the effect of infiltration were detected high heavy metal concentrations. Analyzing the Scholler semi-logarithmic diagram results, fertilizer (ammonium nitrate) has been determined to be effective to heavy metal pollution in groundwater in different three periods. This diagram never before has been used to determine pollutant sources. This study will be a guide for estimating sources of soil and groundwater pollutant in agricultural areas.

Keywords: Göksu delta, groundwater, heavy metal pollution, fertilizer, Schoeller semi logarithmic diagram

INTRODUCTION

With the rapid economic growth and fast population increase, land use has become one of the most important factors responsible for groundwater quality. The agricultural irrigation effluent, wastewater discharge, and domestic effluents have largely contributed to the contamination of groundwater (Aravena et al., 1999; Compton and Boone, 2000). However, due to rapid unplanned urbanization, land use, excessive use of synthetic chemicals in agriculture ,together with overexploitation of the coastal aquifers have collectively resulted in qualitative and quantitative degradation of the groundwater resources in Göksu Delta (Demirel et al., 2011). The lack of delta-wide management strategies and occurrence of pollution from various point and diffuse sources only add complexity of the problem.

The purpose of this study was to use different a method as the Schoeller semi logarithmic diagram to prove the impact of agricultural activity influencing the heavy metal composition of groundwater in Göksu Delta.

Description of the study area

The Göksu Delta is situated in the Mediterranean Sea region of the southeastern part of Turkey and extends from 36°15'-36°25' of latitude north to 33°55'-34°05' of longitude west. The Göksu Delta area is bounded by the Taurus Mountains on the northern side and by the Mediterranean Sea on the southern side. The southern portion of the Göksu Delta area is a delta plain made up of sediments from Göksu River. In the Göksu Delta area, climate is characterized by hot and dry periods in summer and by warm and wet periods in winter, which is typical for the coastal zones around the Mediterranean Sea. The mean annual temperature in this area is 19°C. Showers start in October, and continue till mid April and the maximum rainfall occurs in December. The Göksu Delta area receives slightly higher than 557.1 mm of precipitation annually, and extended periods (i.e., 3–4 11-months) without precipitation are common (SEPA, 2009). Göksu Delta is an internationally important wetland due to its location being on a bird migration route. The Environmental Protection Department of the Ministry of Environment has declared the Göksu Delta as a Special Environmental Protection Zone to protect the area against pollution and exploitation, and to ensure that natural resources and cultural assets have a future. The delta is classified as a Wetland of International Importance according to the Ramsar Convention on Wetlands of International Importance. The Göksu Delta has also a special significance for being one of the few remaining areas in the world where sea turtles (Caretta caretta, Chelonia mydas) and blue crabs (Callinectes sapidus) lay their eggs (Ayas and Kolonkaya, 1996). Agriculture is income source of the more than 80% of inhabitants in delta. Approximately 10,000 hectares of the delta is used for agriculture. Due to the climatic characteristics of the region, in the delta are being cultivated each month of the year. In most products which are citrus fruits, strawberries, tomatoes, wheat, and rice are grown in Delta (SEPA, 2009). Göksu Delta, Special Environmental Protection Area (SEPA) in the figure below, agricultural products map shows the pattern of agricultural products (Figure 1).

Göksu delta, as a result of intensive agricultural activities, the annual fertilizer use is much higher than the national average (Kaçar and Katkat,2007). Göksu delta 500-700 m depth from the surface that consists of alluvial accumulations. This formation, delta aquifer is generally unconfined aquifer. Delta, has more than 1000 wells, but it is not completely alluvial layers. The alluvial aquifer consists of a heterogeneous mixture of gravel, sand, silt, clay and sandy-clay. Akdere Formation contains various rocks with differing compositions including sandstone, siltstone, dolomite and limestone (Demirel et al.,2011) (Figure 2).



Figure 1. Göksu Delta, SEPA, agricultural products map (SEPA, 2009). (blue : Garden crops (citrus, pomagranates, aprikots, tomatoes, peppers, strawberries), orange : Garden crops 2 (wheats, corn, vegetables, peanut, strawberry), pink : strawberry, light green : rice, dark green : cereals, brown: reed-bed).



Figure 2. Map showing the geology of the study area (Demirel et al., 2011).

Accordingly, the hydraulic properties of these deposits can differ greatly over short distances, both laterally and vertically. Heavy metals are mainly introduced into groundwater by agricultural and industrial activities, land filling, mining and transportation. There are various possibilities for the fate and transport of heavy metals in soil and ground water. Solved metal ions Men+ can be taken up by plants, can be sorbed unto mineral phases, or can be bound unto particulate organic matter via complexation/sorption mechanisms. These colloids are very mobile in soil and groundwater systems and will thus increase heavy metal mobility. Dissolved organic matter also plays an important role for heavy metal mobility. The ever growing world population requires intensive land use for the production of food, which includes repeated and heavy input of fertilizers, pesticides, and soil amendments. Fertilizers are added to the soil in

order to provide additional nutrients to crops or by changing soil conditions such as pH to make nutrients more bio available. Pesticides are used to protect crops. Soil amendments are often derived from sewage sludge animal manure, and dredged sediments from harbours and rivers. As soil, surface, and groundwater are closely interconnected systems, metal introduced into soils can also affect aquifers or surface waters by infiltration. Also irrigation may trigger release of heavy metals (Bradl, 2005). However, the heavy metals in the leachate cannot be decomposed by the microorganisms in the soil layer. However, these metals can be concentrated by organisms and have strong mobility, causing pollution to soil and groundwater. Completely removing heavy metals from soils is difficult, thus posing a serious threat to human health (Qiang et al., 2014).

Groundwater quality in a region is largely determined by the natural processes (lithology, groundwater velocity, quality of recharge waters, interaction of water with soil and rock, and interaction with other types of aquifers), anthropogenic activities (agriculture, industry, urban development, and increasing exploitation of water resources) and atmospheric input (Chan ,2001) (Helena et al., 2000). Groundwater maybe contaminated upon leaching of chemicals in the soil surface towards the aquifer. In recent years, due to the use of heavy metals in industry, has increased the rate of people exposure to heavy metals. In addition, significant amount of toxic elements are left by fertilizers and pesticides used in agriculture in soils. The most important of these toxic elements are cadmium, lead, nickel, arsenic, and copper.

Research shows that raw phosphate rock imported from abroad to produce phosphorus fertilizer, were higher in heavy metal contents. Studies have shown phosphorus fertilizer richer than other fertilizers for Cd and As (Köleli and Demir, 2008). Mediterranean region where the most intense greenhouse, a region that is consumed large quantities of fertilizers and pesticides (Kaçar and Katkat, 2007).

In a study which is Hong Kong coastline, rain and groundwater samples were collected in different seasons. Contents of trace elements and heavy metals (Zn, Cr, Cu, Cd, Pb, Fe, Mn, V, Co and Mo) of these samples were investigated in this study. According to studies, Mn, V, Co, Mo ions was determined high concentration. Having studied the seasonal variation; trace elements and heavy metals concentration wet seasonal is higher than dry seasonal (Leung and Jiao, 2006).

In a study, which is grown soil and product in China Pearl River Delta, was researched content of heavy metal of soil and product. Cd and Pb concentration was determined that there is more in soil and paddy (product). Soil which made cultivation paddy is content heavy metal less, because infiltration process is higher than according to other soil. In soil of Pb Zn and Cu ions were connected portion was investigated. Pb widely was connected to the Fe-Mn oxide phase. A significant portion of Cu was detected, the organic sulphides was connected and in the residual phase (Wong et al., 2002).

A study of agricultural soils in Zagrab Croatia, were investigated pollution source of some heavy metal. Researchers with their done statistical studies, showed that pollution of Cd, Cu, Pb, Zn be from anthropogenic sources (agricultural activities) and pollution sources of Fe, Mn and Ni cause by the geological structure of the area (Romic and Romic, 2003).

The purpose of this study was to use different a method as the Schoeller semi logarithmic diagram to prove the impact of agricultural activity influencing the heavy metal composition of groundwater in Göksu Delta.

MATERIAL AND METHODS

Sample collection and treatment

As to reveal of heavy metal pollution in groundwater, three period groundwater Sample was taken in the Göksu Delta.

- 1. Period-February, 2011
- 2. Period-November, 2011
- 3. Period- September, 2012.

Source of heavy metal pollution in groundwater in the Göksu Delta to determine were taken chemical fertilizer sample (ammonium nitrate). Ammonium Nitrate fertilizer is used the most widely in study area. Ammonium nitrate fertilizer is solid form. Ammonium nitrate fertilizer was analyzed form of aqueous solutions. According to "Water Pollution Control Regulations the Sampling and Methods of Analysis" water samples were taken from 58 wells. By addition of acid (HNO₃-Nitric Asid) to one of bottles, were used for heavy metal analyzes. The samples were stored 4°C in the refrigerator at until analyzed. The sample points (wells) are shown in Figure 3, The depths of sampled wells are 40–150 m, which most of them penetrate the alluvial aquifer.



Figure 3. Groundwater sample points in Göksu Delta.

Analytical procedures

All analyses were carried out at the Mersin University Environmental Engineering Department, Mersin, Turkey. Measurements of Electrical Conductivity (EC), Salinity (Sal), Temperature (T) and pH were made in the field using a pH/Cond 340i WTW meter. Groundwater samples obtained at the point of coordinates (x, y, z) Global Positioning System (GPS) was measured with Magellan Explorist in place. Heavy metal concentrations of the groundwater samples (Iron (Fe), copper (Cu), chromium (Cr), manganese (Mn), nickel (Ni), Zinc (Zn), Cadmium (Cd) and lead (Pb)) were analyzed with Agilent model 7500 Inductively coupled plasma-mass Spectrometry (ICP-MS) which situated in the Central Laboratory of Mersin University. In this study were interviewed District Directorate of Agriculture technicians, farmers and pesticide dealers. As a result of these talks were created following table for the use of pesticides and

fertilizers on the delta (Table 1).

Table 1. The most commonly used fertilizers and pesticides by months in Göksu Delta.

						_						
Commonly used	1	2	3	4	5	6	7	8	9	10	11	12
fertilizers and												
pesticides												
NH ₄ -NO ₃									*	*	*	
NH4-SO ₄								*				*
Phos phorus Fertilizer									*	*	*	
FeSO4							*				*	
Fetrilon combi-2												
(Mn-Fe-S-Zn-Cu-Mo-												
B)					*							
Active ingredient:												
Abamectin 18g/L-												
TORPEDO EC					*	*						
Active												
ingredient:acetonitrit-												
HEKPLON 20 SP												
(insecticide)					*	*						
Active												
ingredient: pyri proxfen-												
100gr/L-ADMİRAL												
10EC							*					
Iron fertilizer –consist												
of Ferrostrene-% 4.8												
ortho-ortho.			*	*								
TORPEDO-												
(insecticide) +												
DOMARK (fungicide)			*	*	*							
KODEFOL 680												
(consist of phosphorus)			*	*	*							
Chlorine profosel ethyl						_						
(ne matoci de)							*					

RESULTS AND DISCUSSION

Groundwater and fertilizer sample minimum/maximum heavy metal values of wells each period are shown in the following table (Table 2).

Table 2. Shown minimum/maximum heavy metal values of wells each period.

	1.Period		2.Period		3.Period	
Concentration mg/L	Min.	Max.	Min.	Max.	Min.	Max.
Fe	0.0000	3.5200	0.0000	1.4100	0.0011	1.1300
Mn	0.0010	0.7600	0.0000	0.2885	0.0001	0.2160
Cu	0.0000	0.0030	0.0000	0.0002	0.0000	0.0016
Cr	0.0000	0.0040	0.0000	0.0055	0.0000	0.0070
Ni	0.0000	0.1320	0.0002	0.0092	0.0005	0.0300
Zn	0.0000	4.4600	0.0002	2.1700	0.0017	1.9490
Cd	0.0000	0.0338	0.0000	0.0061	0.0003	0.0110
Pb	0.0000	0.0105	0.0000	0.0081	0.0020	0.0280

*0.0000 : below detection limit

Minimum/maximum (pH, Temperature, Electrical Conductivity, Salinity) values of wells each period were shown with Table 3.

Table 3. Minimum/maximum pH, temperature, electrical conductivity, salinity values of wells each period.

	1.Period		2	.Period	3.Period		
Pa ramete rs	Min.	Max.	Min.	Max.	Min.	Max.	
pH	7.12	8.47	7.2	8.7	7.10	8.34	
Temperature (°C)	15.1	21.7	16.3	21.7	21.10	29.8	
Electrical Conductivity (μS/cm)	459	5240	451	5600	483	5830	
Salinity (sal)	0	2.8	0	3	0	3.2	

The pH of groundwater showed alkaline characteristic caused by geological structure. Water temperature gradually increased toward the warmer seasons. Wells which are near the coast were showed high Electrical Conductivity. Values of salinity have increased in parallel with the Electrical Conductivity (Table 3).

Results of the three periods heavy metals in the groundwater were compared with the limit values TC. Water Pollution Control Regulation (WPCR) and US. Environmental Protection Agency (EPA). The following tables (Table 4, Table 5) shown wells of exceed the limit value.

	0		
Concentration mg/L	1.Period	2.Period	3.Period
Fe	D1,D9,D21,D20,D16,D2,B6,B10	D21,D20,D16,D15,B6	D1,D16,D14,B14,B5,B7,B9
0.3 mg/L			
Mn	D1,D21,D22,D19,D20,D16,D2,D3,	D21,D19,D17,D2,D6,D7,	D19,D2,D7,B14,B7,B10
0.1 mg/L	D7,D9,D13,B26,B13,B14,B4	B14,B7,B11,B10	
Cu			
0.02 mg/L			
Cr			
0.02 mg/L			
Ni	D16,B26,B14		D7
0.02 mg/L			
Zn	B26,B14,D16	B14,B26,B19,B20,	D2,D5,D8,B28,B26,B19,B20,
0.2 mg/L		D23,D19,D5	B14,B7,B17
Cd	D17,18,B26,B29,B5,B32	D17	B7,B33,B32
0.003mg/L			
Pb		B14	B24,B12,B32,B33
0.001 mg/L			

Table 4. Wells exceeding the WPCR -1.class water limit values.

*-----: Do not exceed the limit value

Table 5. Wells exceeding the EPA limit values.

Concentration			
mg/L	1.Period	2.Period	3.Period
Fe			
no limit values			
Mn	except(D10,B25,B21,B8,B3),all	except(D23,D24,D4,D25,D10,B25,	D23,D24,D25,D10,B26,
0.0015 mg/L	wells	B28,B31,B8,B3,B32),all wells	B28,B31,B17,B8
Cu			
1.13 mg/L			
Cr			
0.02 mg/L			
Ni			
0.02 mg/L			
Zn			
0.2 mg/L			
Cd	D17, D18, B26, B29, B5, B32	D17	B7,B33,B32
0.003mg/L			
Pb			B24,B2,B18,B32
0.001 mg/L			

*-----: Do not exceed the limit value

Mostly the wells (called D code) in the east of the Delta have been found to exceed the limit values. The reason of heavy metal pollution than other regions may be due to intense settlements and agricultural activities in the eastern of delta. Although 1.Period iron concentration are less

than the other periods, the maximum concentration was found to be 1.Period. A dense region of the iron ions was evaluated with Figure 1 and was found that these regions are corresponds to citrus fruits, strawberries, rice production area. According to the iron results with comparison the amount of fertilizers and pesticides used in the delta (Table1); FeSO4 for the production of citrus in November, iron fertilizer for the production strawberries in March, have been used. These results explain increase of the amount of iron in the wells. In all periods Cr, Cu, Ni pollution were not detected in groundwater. Zinc is found chemical fertilizers and agricultural chemicals. In Turkey soils is a significant level of zinc deficiency. This also, increased the use of containing zinc fertilizer in Turkey. Sources of cadmium in agricultural soils are industrial activities, phosphorous fertilizers, wastewater and atmospheric deposition (Assche and Clijsters,1990). In terms of lead and cadmium, has not been found in excess of the limit values of wells.

In a study which is agricultural soils in Göksu Delta, was investigated heavy metal pollution (As, Cd, Pb, Cr) from agricultural sources. Results have showed that due to excessive and irresponsible use of fertilizer and pesticides in the Göksu Delta, the delta soil exceeds the limit values (Köleli, N. & Demir, A. 2008). Soil properties, such as pH, clay and organic matter (OM), are the main factors that impact heavy metal mobility (Guannan et al.,2014). Soil OM can bind with heavy metals creating stable states (Neagoe et al.,2011).

Metal ions can be retained in the soil by sorption, precipitation, and complexation reactions, and are removed from soil through plant uptake, leaching, and volatilization. The fate of metals in the soil environment is dependent on both soil properties and environmental factors. Retention of charged metal solute species by charged surfaces of soil components is broadly grouped into specific and non-specific retention (Bolan et al., 2003; Sparks, 2003). Both soil properties and soil solution composition determine the dynamic equilibrium between metals in solution and the soil solid phase. The concentration of metals in soil solution is influenced by the nature of both organic and inorganic ligand ions, and soil pH through their influence on metal sorption processes (Bolan et al., 2003; Harter and Naidu , 1995). Other chemical interactions that contribute to metal retention by colloid particles include complexation reaction between metals and the inorganic and organic ligand ions. As might be expected, the organic component of soil constituents has a high affinity for heavy metal cations such as Cu, Cd, and Pb because of the presence of ligands or groups that can form chelates with metals (Harter and Naidu ,1995). The organic amendments can directly or indirectly alter the distribution and availability of metals in soil (Walker et al., 2004; Bernal et al., 2007). The effect of organic residues in the mobility and bioavailability of metal (loid)s in soils depends on the specific metal, soil type, and the characteristics of the amendment (EC, CEC, pH, and humification degree) (Walker et al., 2004; Walker et al., 2003). For instance, there are reports that amending contaminated soils with compost may increase the mobility and leaching of metals (Clemente et al.,2010). Previous researches for the Göksu Delta soil properties were determined pH 8 (alkaline), 25% CaCO3, 2% organic matter, 30% sand, 26% clay and 34% silt (Kölleli and Demir, 2008). By this information, the low organic matter content of delta soils could be explain the heavy metal ions transport of groundwater from soil.

Using AquaChem program for each period data was transferred Schoeller semi-logarithmic diagram. Schoeller semi-logarithmic diagram is quite often used to comparison similar or different waters. This diagrams show to more than one of ion in a single diagram. In this study Schoeller diagrams were created to aim to show heavy metal concentrations of groundwater which affected of fertilization activities (Figure 4).





When Schoeller diagrams viewed for three period of heavy metal concentration, it created curves for heavy metals concentration of groundwater and fertilizer was observed that are parallel to each other. This parallel curves were shown, when compared in terms of iron, manganese, nickel, zinc ions groundwater were affected by chemical fertilizer.

CONCLUSION

Analyzing the results of three periods of heavy metals in wells water; in 1.period has reached maximum concentration of all metals and to 3. Period has been determined that the correct concentration decreases. Starting from summer months toward autumn, was determined fertilizer and pesticide use correctly intense increase in Delta (Table 1). In the studies in the lands of the Göksu Delta have been found low organic matter content of the delta soil (SEPA,2009). Heavy metals are kept by binding to the functional groups of soil organic matter in the soil. Due to the lack of organic matter in the delta soil will not be able to keep heavy

metals in soil. Heavy metals which taken during the winter season in the samples of first period, it was based on heavy metals accumulated in soil in spring and summer. At the end of this phenomenon heavy metals in soil are transported towards the underground through rain water. The heavy metal characteristics of groundwater were investigated to understand the agricultural factors affecting groundwater quality in the Göksu Delta. The study results showed that agricultural activities were major factor for the heavy metal compositions of groundwater.

This study demonstrated that the Schoeller semi-logarithmic diagram is an effective tool to characterize the agricultural activities effect of groundwater heavy metal components. The Schoeller semi-logarithmic diagram results combined with land uses and fertilization periods helped to distinguish agricultural sources affecting groundwater quality for heavy metal in the Göksu Delta groundwater system.

The results from this analysis will not only help to understand the cause of groundwater heavy metal contamination by agricultural activities in the Göksu Delta groundwater system but also shed light on the methodologies that can be effective to study groundwater other chemical characteristics and identify the dominance of different contributing sources.

So far Schoeller semi-logarithmic diagram is used to determine the source of groundwater. In this study was showed that using Schoeller semi-logarithmic diagram can be used in the comparison of specific pollutant sources of groundwater quality.

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