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Comparative Study on Heavy Metal Contents in Sub-urban Agricultural Fields Irrigated with Sewage and Canal Water of Bahawalpur City, Pakistan

[1]Haroon Rasheed, [1]Karamat Mehmood, [2]Muhammad Farrukh Nisar and [2]Farrukh Jaleel

[1]Department of Chemistry, The Islamia University of Bahawalpur, Baghdad Campus, Bahawalpur (63100), Pakistan
[2]College of Bio Engineering, Chongqing University, Chongqing (400044), China

Abstract: Industrialization results in urbanization, mainly due to efflux of population from rural areas, which often creates different kinds of pollution. The explosion of urban populations in limited resources may lower the life quality and severely imbalance the economy of country. Current study was done in view of various such reports on increasing pollution problems in industrial areas, to analyze the status of heavy metals in irrigation water in Bahawalpur district, Pakistan. Two study sites were selected in the periurban areas of Bahawalpur city (site 1 Village Bindra and site 2 Jhangiwala) and collected the water samples for qualitative analyses including physical characteristics, ionic concentration (meql^{-1}) and heavy metals (As, Pb, Hg, Ni) (ppm). By comparing the national and international standards, it was found that, pH is in the permissible limits. Electrical conductivity (EC), Cation Exchange Capacity (CEC) and Organic Matter (OM) level are higher in top soil layers and become less as gone down to the deep soil layers irrigated with sewage and canal waters. Sodium a dsorption ratio (SAR) values were higher in deep soils and make soils saline sodic giving clayey loam to calcareous texture. Pb, Ni and As were observed in safe limits except Hg which was higher in top and deep soil layers. It is recommended that, a proper treatment of waste water should be done to avoid higher accumulation of Hg in agricultural fields and ultimately food chain.

Key words: Heavy metals · Pollution · Sewage water · Canal water · Physico-chemical

INTRODUCTION

Increase in industry given a population efflux to urban areas that produced the problem of various kinds of pollutions (noise, air, soil and water), which not only bring a decline in life standards but also disturbed economy of local as well as national businesses [1]. When biosphere experienced any non-demanded change in its physical or biological characters, the phenomenon is called as pollution, where the different materials like vehicle exhausts, smoke, dust, smog, metal particles, heat and radioactive materials exceeded the prescribed levels [1]. Various industries, particularly fertilizer, petrochemical, pharma, oil, ghee and thermal power generation houses generate heat and waste into the cultivated fields, water bodies or in to the air in form of either toxic gases or waste effluents. The later mainly contains various byproducts of organic or inorganic nature generated at various stages of productivity often complexes with the heavy metals [2]. In developing countries, there is a poor system or even altogether no system for the disposal of household as well as industrial wastes, rather which is being used to irrigate the agricultural lands. Heavy metals along with some other dangerous substances get their way to accumulate in plant via these industrial effluents and finally approach the food chain to harm human health [3,4].

Toxic elements containing wastewater is under use in irrigation may possibly get entry in the food chain to raise the level of salts through which soil chemistry get disturbed and ultimately alter the yield of crops [5]. Long term use of wastewaters is hazardous for soil as it may deteriorate the soil structure. The crop physiology, growth and yield are affected by direct use of untreated wastewater containing higher level of potassium (K), phosphorous (P) and nitrogen (N) [1]. High total

Corresponding Author: Farrukh Jaleel, College of Bio Engineering, Chongqing University, Chongqing (400044), China.
Tel: +92-3137591002, +86-18716444840.

dissolved solids (TDS) and heavy metals get deposited in the soil and affect the soil badly with the course of time change the soil chemistry which lead to the poor growth of vegetables and crops [6].

Irrigation practices with waste waters contributing significant levels of heavy metals in the soils [7] and creating serious problem to the non irrigated lands by leaching. After the death and decay, plants return the tissue bound heavy metals to the soil thus enrich the soil with the pollutants i.e. heavy metals [1]. Soil residual metals uptake by the pants, depends on various factors such as their solubility, pH of soil, life history of plant growth, kind of plant species, fertilizer contents and soil texture [8]. Plant species developed certain stress tolerant cellular mechanisms by which they can adjust themselves to remove and accumulate heavy metals. Some plant species may accumulate specific heavy metals, causing a serious threat to human health when consumed as a food [9].

Sewage disposal is a serious issue, which often drained to the agricultural fields to irrigate crops and vegetables. The sewage effluent is loaded with organic matter (OM) and other nutrients along with elevated level of heavy metals (Fe, Mn, Cu, Zn, Hg, Pb, Cr, Ni, Cd and Co) when reaching to the cultivating fields [10]. The food chain gets polluted when vegetables absorb heavy metals from soil, immediate polluted air or irrigation waters, that lead to the dietary pathways through uptake of heavy metals by the plants. Heavy metals hardy biologically degrade and goes on accumulation in human body, which may cause illnesses depending on degree of exposure i.e. severe or chronic exposures [11]. On the other hand, trace amounts of heavy metals are also needed in cellular metabolism by our bodies but to certain and recommended limits. The estimated level of heavy metals must not be greater than 5mg m^3 [12]. The spread of heavy metals is well upto a certain limit these days in the developing world particularly, in the environment and local floras. Anthropogenic activities affect the environment and hence more and more heavy metal contamination cases come into knowledge including industry and agricultural practices [13-15]. Majority of the heavy metals are essential part of the plant metabolisms to secure normal growth as co-factors of many enzymes and some proteins. But high concentration in soil of both essential and non essential heavy metals leads to toxicity symptoms that check the growth of plants with declined yield [16]. The higher amounts are the markers for toxicity symptoms and due to their interaction during cellular metabolisms [17].

The pollution problem is increasing day by day in Pakistan due to increased urbanization and trend towards industrialization. In addition, high goods transportation, deforestation, pesticide and insecticide application on all kinds of cultivations plants i.e. vegetables, fruits and different crop plants. In a polluted environment, air get customized to high concentration of different lethal gases carrying many of the heavy metals, where the later gets its way to food chains through various sources ultimately reach human bodies to cause some toxic effects. Lead, copper, mercury and chromium are enormously under use in industry [18]. The role of Hg and Pb have not been established to be essential for either plants or animals [5]. The above discussion led us to study levels of heavy metals in purely agricultural city of Bahawalpur, where the city effluent is mostly in irrigated usage in different vegetable fields irrigated with sewage effluents and canal water.

MATERIALS AND METHODS

Study Area: Bahawalpur is an agricultural district situated in the south of Punjab Province, Pakistan. In Bahawalpur, the number of industrial units is very small but their effluent is also used for irrigation purpose. About 72.7% rural populations involved in agricultural practices, while rest lives in urban areas. Mostly the area is canal irrigated but the suburban areas are also prone to waste and sewage water flooding for cultivating different vegetables and forage for grazers. We selected two study sites i.e. Village Bindra (study site 1) and Jhangiwala (Study site 2), which are the major producers of vegetables for the local city demands.

Sample Collection and Analysis of Samples:

Sewage Effluent: The sewage effluent used for irrigation purpose was taken from the field of selected area where vegetables were irrigated with it. These samples were collected in the month of November, 2009. These samples were well mixed to achieve uniform composition. Samples for heavy metals determination were also collected. Nitric acid (5ml) was added to each sample to preserve samples till analysis. Clean plastics bottles were caped and air tight with plastic tape, labeled date wise accordingly and were taken back to the laboratory for further analyses. Position of each sampling point was recorded by GPS (Global Positioning System) for future reference.

Analytical Methods: All chemicals and reagents were of the analytical grade and were obtained from BDH Chemicals Ltd, UK. Working standards were prepared from Certified Reference Material (CRM) with certificate of analysis traceable to National Institute of Standards and Technology (NIST). Analytical techniques followed during analysis along with instrument make, model and method reference are mentioned in Table 1. Sodium adsorption ratio (SAR) and residual sodium carbonate (RSC) were calculated using different equations [19]Richards, 1954). Samples were filtered with Whatman filter paper No 42 and were analyzed to determine Co^{2-}_3, HCO^{1-}_3 Cl^{1-}, SO^{2-}_4 pH, EC (Electrical Conductivity) Na^+, K^+, Ca^{+2} + Mg^{+2} and Ca^{+2}. Residual sodium carbonate (RSC) and Sodium adsorption Ratio (SAR) were also computed for samples.

$$SAR = Na^+/[(Ca^{+2} + Mg^{+2}) /2]½$$
$$RSC = (CO_3^{-2} + HCO_3^-) - (Ca^{+2} + Mg^{+2})$$

Magnesium adsorption ratio (MAR) was calculated according to Raghunath [20].

$$MAR = Mg^{2+}×100/Ca^{+2}+Mg^{+2}$$

All cations and anions concentrations are expressed in meq L^{-1}.

Heavy Metals Analysis: Analysis for four major heavy metals, i.e. Ni, Pb, Hg and As were performed on Atomic Absorption Spectrophotometer (AAS) (Analytic Jena Germany) equipped with D2 background correction lamp was used. The results obtained from the AAS analysis of ground water samples for selected area indicated that values of Cr, Ni, Pb, Co and Cu were below detectable limits and were not detected (ND). The values for physico-chemical (pH, EC, SAR) and chemical properties (Na^+, CO_3^{-2} + HCO_3^{-1}, Ca^{+2} + Mg^{+2}) were calculated by using the standard procedures and averages were made by using Microsoft Excel(R).

RESULTS AND DISCUSSION

The present study was conducted in order to evaluate the effect of municipal sewage on soil. The transportation of heavy metals from sewage water to soils which are being irrigated with sewage water of Bahawalpur city and which are being used for cultivation of various crops especially vegetables to supply in local market. Mean while site was also selected for canal water irrigation (Study site 2). The quality of sewage water and canal water was evaluated for general irrigation water quality parameters and then for heavy metals. Later the soil chemistry through similar analyses were studied to evaluate the level of heavy metal contents and compared with the national and international standards. Comparative chemistry of both sewage and canal water is given in Table 1.

The soil with a pH <8.5 is termed as sodic soil and phenomenon as sodicity. The pH of soil at 0-6 cm depth taken from village Bindra ranged from 7.9-8.1 in the different vegetable fields, while at Jhangiwala fields pH ranged from 7.9-8.1 (Table 1). Overall a fluctuating or irregular trend in the increase or decrease of pH at various depths was noted (Table 1). The pH in all the samples from all fields was found within the international permissible limits i.e. 8.5 for various agricultural crops and vegetables (Table 1). However the soil receiving canal water had a little bit low pH of 7.93 (Table 1). In general the pH was not problematic for sewage water irrigated areas because of organic matter (OM) regulation in the sewage water as it contained < 20 % of OM [21]. In canal water irrigating fields the pH was low due to calcareous nature of soil and canal water which contain a lot of calcareous mineral water.

The EC of soil reflects the extent of salinity and soils having EC <4dSm^{-1} are characterized as saline soils [22]. The EC of soil irrigated with city effluent ranged from 1.8-9.8 dSm^{-1} at the depth of 0-6 cm, while at 6-12 cm depth, it ranged from 1.3-6.5 dSm^{-1} and there is a declining trend (Table 1). The upper soil stratum (0-6 cm) have higher EC than deeper ones (6-12 cm), might be due to salts in effluent applied for cultivating crops. The lower EC values of soil in the lower horizon advocates the development of soil from normal parent material was ranged from 5.1-8.5 dSm^{-1} respectively and were categorized as saline. The EC of soil receiving canal water ranged from 1.0-1.41 dSm^{-1} (Table 1), because the salts in canal water are low so the EC in soil receiving this water is also low.

Organic matter (OM) improves the physical and chemical properties of soil and is a ready source for essential minerals to plants. The organic matter contents of soils ranged from 0.34- 0.61 % in the surface soil layer (Table 1). Further, in deeper soil layers the OM contents were decreased and previously Bohn *et al*. [23] reported that organic matter was decreased with increasing soil depth. The high organic matter contents in the field receiving sewage irrigation water were due to its addition in sewage effluent. More over the intense cultivation restored the soils as crops continuously add plant residues and litter in the soil which is considered as a significant source of soil organic matter [24].

In spinach fields, soil receiving waste water, the SAR was ranged from 2.61-4.35-3.00-6.37 in top soil (0-6 cm) and in deep soil (6-12 cm) respectively, while the fields receiving canal water, the SAR value ranged from 5.27-5.37, at the above mentioned depths of soil. The cauliflower fields which receive waste water has SAR value ranged from 4.45-5.61 in the upper layer (0-6 cm) of soil and 3.49-6.00 in deep soil (6-12cm) layers, but for canal water cauliflower fields the SAR was in 6.30-5.90 range. The SAR value for the brinjal fields receiving sewage water, was 3.59-5.65 in top soil (0-6 cm) and 3.69-4.86 for deeper soil (6-12 cm) and for the canal water SAR values were 5.08-5.00 in the top and deeper soil layers (Table 1). Based on the SAR values for various fields, it is seen that mostly the soil is of saline sodic in nature. In general the SAR decreased towards the depth i.e. the sewage water irrigation (EC, 0.935-1.5l2 dSm^{-1}, SAR 4.0-6.44, RSC 0.51-7.18 mmol L^{-1}) in spite of all the former management, has shown its impact on soil salinity. Although at the observed levels of EC, SAR, RSC and pH, most of the crops could yield adequately because of their potent tolerance level as previously mentioned by different workers [25]. Hence, it is proposed that for such irrigation waters, the formers must follow some management practices, like use of Ca source, or organic matters [26,27] and any other available and suitable method described in literature [28].

All the soils investigated, were clay loam except the soil receiving canal water were loam and two samples of soil receiving waste water (Table 3). This clay loam texture is expected not to impede the internal drainage of soils [29]. This fact is also reflected in the form of very low EC and SAR. In addition, all the soils were slightly calcareous up to the upper layer and were moderately calcareous in the lower surface. This pattern of lime distribution also indicates the good internal drainage due to which under extensive and heavy sewage water irrigation, the lime moved towards lower soil layer. Moreover, higher OM in the upper layer tended to decrease the formation and accumulation of lime in these soil depths. It is well established that OM decomposition helps to convert $CaCO_3$, into more soluble $Ca(HCO_3)_2$ [30] and this indicates better movement of this compound deep in soil. The calcareous soil is classified as the world best buffer soil because any thing we add into these soils, the alteration is for the time being and after some time the soil revert into its normal condition.

Cations exchange capacity (CEC) represents the capacity of soil to hold the exchangeable cations i.e. greater the CEC greater the fertility of soil. The CEC of top soil layer (0-6 cm) ranges from 20.86-9.56 $meqL^{-1}$. while for deeper layers (6-12 cm) it ranges from 20.34-11.34 $meqL^{-1}$ (Table 1). In canal water irrigated fields had CEC ranged from 10.42-13.90, the higher value of CEC confirms the higher concentration of organic matter in soil receiving city effluent than that of soil receiving canal water. Since organic matter carries very high concentration of negatively charged ions [23].

The Pb concentration in the top soil (0-6cm) ranged from 0.3541-8.181 mg/kg^{-1} and decreased by increasing soil depth (Table 2). The critical limit for Pb in soil is set to 500 mg/kg^{-1} [31]. Thus Pb noted within the permissible limits in all the soil samples when compared with the national standards [32]. Various concentration of Pb has been reported to be varied according to the irrigation practices with sewage waters [3,33,34]. Ghafoor *et al*. [4] evaluated through a lysimeter experiment that Pb was very less mobile element in the soil and thus tended to accumulate in the surface layers of soil when added with water application and confirmed the previous reports [22] even in the sandy, acidic, OM deficient and high precipitation receiving soils. The Pb is also used in automotive fuels and gasoline, released into the environment consequently gets its way to the surface of soil by deposition in form of complexes [35]. The Pb in soils irrigated with canal water was in lower level i.e. ranged from 6.163-8.588 mg/kg^{-1}, than the permissible limits of Pb in soil when compared the results with the national limits [32].

Nickel (Ni) in the top soil (0-6 cm) ranged from 0.02462-0.8269 mg/kg^{-1}. This concentration is less than the critical limits of 20 mg/kg^{-1} [31] and the concentration of Ni in soil irrigated by canal water is ranged from 0.2452-0.4435 mg/kg^{-1}. It was observed that the concentration of Ni decreased towards soil depth (Table 2). The higher concentration of Ni in top soil layer might be due to the addition of Ni through the application of sewage water containing high metal concentrations as previously reported for cauliflower, spinach, tomato and barseem [36]. This distribution of Ni could also be due the fact that higher OM in surface layers complexes to retain Ni in upper soil stratum [23], in spite of the good drainage as indicated by medium type soil texture and low increase in EC and SAR over a long period of sewage water irrigation. Thus it is natural to have higher Ni concentration in the top soil layers.

Table 1: Physico-chemical properties of soil receiving sewage effluent and canal water Bahawalpur.

		Sewage Water Canal								
		Spinach Field		Cauliflower Field		Brinjal Field		Water		
Sr. No.	Parameters	0-6 cm	6-12 cm	0-6 cm	6-12 cm	0-6 cm	6-12 cm	Spinach Field	Cauliflower Field	Brinjal Field
1	pH	7.9+1.08	7.9+1.08	7.96+0.05	8.0+0.05	8.03+0.11	8.1+0.01	8.0+0.141	7.95+0.212	7.90+0.282
2	EC dSm^{-1}	6.66+4.119	4.60+2.193	2.1+0.2	1.73+0.153	2.56+0.305	2.43+0.351	1.25+0.07	1.1+0.141	1.15+0.07
3	OM (%)	0.61+0.017	0.46+0.017	0.36+0.023	0.55+0.035	0.50+0.04	0.34+0.005	0.425+0.035	0.43+0.028	0.44+0.041
4	SAR	3.25+0.952	5.10+1.83	5.09+0.589	4.73+1.255	4.28+1.180	4.22+0.591	5.310+0.06	6.1+0.282	5.04+0.06
5	CEC $molKg^{-1}$	16.6+2.07	16.9+2.67	16.25+2.99	18.66+1.65	12.46+2.66	13.38+1.87	14.09+0.268	11.41+1.40	12.25+1.343
6	Lime (%)	46.48+6.91	55.99+8.25	58.08+3.94	61.40+3.58	56.96+1.57	53+9.44	54.30+1.05	62.25+1.06	57.1+3.53
7	Saturation (%)	56.00	56.67	46.00	53.67	47.33	45.33	44.00	52.00	38.00
8	HCO^{-3}	1.03	1.27	1.266	1.366	1.66	2.00	0.9	1.2	1.6
9	Cl^{-1}	2.516	3.066	7.7	4.06	3.066	3.00	4.4	3.5	0.4
10	SO_4^{2-}	0.666	0.693	0.633	0.600	0.626	0.573	0.31	0.28	0.10
11	Na	5.286	2.546	2.85	2.43	2.456	3.07	1.21	0.69	0.05
12	K	7.946	6.750	4.016	3.493	2.930	3.013	0.12	0.13	0.05
13	P	19.376	15.7	5.566	6.736	6.266	4.300	6.9	6.9	6.3

Table 2: Concentration of heavy metals in soil irrigated with sewage effluent and canal water Bahawalpur.

		Sewage Water								
		Spinach Field		Cauliflower Field		Brinjal Field		Canal Water		
Sr. No.	Parameters	0-6 cm	6-12 cm	0-6 cm	6-12 cm	0-6 cm	6-12 cm	Spinach Field	Cauliflower Field	Brinjal Field
1	Pb	0.511+0.242	0.458+0.186	3.128+1.184	3.128+1.184	7.583+0.510	6.160+0.614	0.180+0.024	6.158+0.776	8.643+0.078
2	Ni	0.70+0.155	0.483+0.252	0.439+0.024	0.45+0.212	0.193+0.157	0.214+0.143	0.433+0.013	0.245+0.014	0.230+0.02
3	As	0.036+0.005	0.141+0.004	0.043+0.005	0.174+0.230	0.044+0.005	0.048+0.004	0.025+0.007	0.019+0.001	0.02+0.01
4	Hg	0.066+0.02	0.099+0.009	0.027+0.023	0.088+0.017	0.061+0.05	0.075+0.06	0.049+0.007	0.039+0.024	0.018+0.001

Table 3: Texture of soil receiving sewage effluent and canal water Bahawalpur.

		Sewage Water								
		Spinach Field		Cauliflower Field		Brinjal Field		Canal Water		
Sr. No.	Parameters	0-6 cm	6-12 cm	0-6 cm	6-12 cm	0-6 cm	6-12 cm	Spinach Field	Cauliflower Field	Brinjal Field
1	Clay (%)	29+1	34+1.73	30+9.53	33.33+1.52	34.33+3.21	36.76+2.08	21+1.41	22.5+3.53	35+4.24
2	Silt (%)	42.33+11.23	44.66+8.02	43.33+7.63	42+2.64	3.33+5.83	46.76+5.50	41.5+2.12	32.5+3.53	34+5.77
3	Sand (%)	28.66+10.67	21.33+10.01	26.76+1366	21.33+3.05	32.33+5.50	16.76+4.16	42.5+3.53	34.5+5.77	35.5+3.53
4	Texture Class	Clay Loam	Clay Loam	Clay Loam	Clay Loam	Clay Loam	Clay Loam	Loam	Loam	Loam

The concentration of Arsenic (As) in soil receiving city effluent was ranged from 0.03-0.05 mg/kg^{-1} in the surface layer of soil and the concentration of As increases with soil depth and was noted in the safe limits of soil i.e. 30 mg/kg^{-1} [31]. The concentration of As in soils irrigated with canal water was ranged from 0.01-0.02 mg/kg^{-1}, were also in the safe limits. Waste water used for irrigation of soil contained high concentration of As in it and is not likely to be dissolved or washed out by flooding with water or not oxidized due to its affinity with Fe, Mn, Al and other minerals in soil. As get accumulated in the upper soil stratum (0-6 cm) and noted >83 mg/kg^{-1} [37], while in deeper soil layers the concentration range was 4-8.0 mg/kg^{-1}. As also used in pesticides, insecticides and in fruits ripening hormones or chemicals, all these sources led the As to enter into soil and rises concentration in soil. Leo and his colleagues [38] reported that the As used into the pesticides and insecticides are the major sources of soil contamination with excess of As, though due to waste water, sorption of As with Fe and Al, it mainly restrict in the upper soil stratum. Inspite of occurrence in upper soil layers it remained hardly available to the plants because As does not leach deeper into the soil so deeper roots failed to uptake too much As.

The concentration of Mercury (Hg) in soil irrigated with city effluent was ranged from 0.00-0.08596 mg/kg^{-1} in upper soil layers (0-6 cm). The concentration of Hg was not in the safe limits than the permissible limits in soil i.e. 1.0 mg/kg^{-1} as per Alloway's [31] given standards. The concentration of Hg increases with the increase of soil depth i.e. from 6-12 cm. The source of Hg in soil might be the pesticides and insecticides, the farmers used to protect different crops and vegetables against insects, pests and rodents, also aid in Hg depositions into soil besides waste waters used for irrigation purposes. The waste water contains high level of OM in it, which contain large number of heavy metals in it that make their way by the process of lechates into deeper soil layers [39]. In our results, we have observed that the concentration of Hg increases along the depth of soil. As the mercury volatilized into other forms after the extraction of Hg from soil by DTPA solution Conc. HNO_3 to prevent the volatilization of Hg by lowering the pH of the extract as the solubility of the Hg increased with reducing pH [40](NRCS 2003). Also the same Hg volatilization was also reported by various micro-organisms [40].

By comparing the national and international standards [41,42], it was found that, pH is in the permissible limits. EC, CEC and OM level are higher in top soil layers and become less as gone down to the deeper layers of soils irrigated with city effluent and canal waters, on the contrary the SAR values increased in contrast to both EC and OM along the depth of soil. Based on these SAR values, it was observed that soils are saline sodic in nature with clayey loam to calcareous texture and due to the later property, such soils are considered as the best soils in view of buffering to maintain the medium static. For the heavy metals like Pb, Ni and As, all these were observed in safe limits and the concentration decreased in deeper soil layers of both sewage and canal water irrigated soils. In both sewage and soil water irrigated soils, Hg was higher in top and deeper soil layers and over the safe limits set by national and international standards. The higher accumulation of Hg was due to the over utilization of pesticides, insecticides etc.

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