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Effect of Fertilizer Application on Paddy Soil Heavy Metals Concentration and Groundwater in North of Iran

[1]*Hajar Boudaghi Malidareh,* [1,2,3]*Amir Hossein Mahvi,* [1]*Masoud Yunesian,*
[1]*Mahmood Alimohammadi and* [1]*Shahrokh Nazmara*

[1]Tehran University of Medical Sciences, Tehran, Iran
[2]Center for Solid Waste Research, Institute for Environmental Research,
[3]Tehran University of Medical Sciences, Tehran, Iran

Abstract: Fertilizers contain heavy metals that can cause serious problems in water and soil. Contents of heavy metals in water and soil are very important because water and soil are the critical first link in the food chain. The purpose of this study was to determine the effect of fertilizer application on paddy soil heavy metal concentration and groundwater in North of Iran.20 composite samples of paddy soil were taken before and after fertilization at 0 to 30 cm depth and from groundwater. Also, chemical analysis of composite samples of three fertilizers with high consumption was performed. As, Cd and Pb were extracted by acid digestion method in water and fertilizer samples were extracted and measured by inductively coupled plasma-optical emission spectrometer. Cd concentration in Triple Super Phosphate (TSP) was higher than standard California Department of Food And Agriculture (CDFA). A significant relationship between the amount of potash fertilizer and cadmium concentration in soil was observed ($p<0.05$). The highest concentration of Cd in Phosphate fertilizer was due to the lands that used more potash fertilizer than usuall consumption also Heavy metals in water were lower than detection limits. No relationship between the amount of heavy metals in the soil with water and heavy metals in the fertilizer with water was observed. Although concentration of heavy metals in soil and water has not exceeded before and after fertilization in this study, but frequent use of chemical fertilizers and pesticides in common agricultural fields can cause increase in rate of heavy metals in soil and water, So its expected health authorities study annual level of heavy metals in soil and groundwater.

Key words: Chemical Fertilizer · Heavy Metal · Paddy Soil · Groundwater

INTRODUCTION

Due to growth in population, exploitation of water and soil resources and high consumption of fertilizers and pesticides can cause chemical pollution. Applying chemical fertilizer and pesticides reduces the quality of underground waters. The basic idea of farmers in using chemical fertilizer is achieving better performance and more products [1]. Chemical materials gained out of agricultural activities cause serious pollution of surface water and groundwater in many countries [2]. Here are some factors which can lead to gathering of heavy metals in soil: disposal of municipal and industrial wastes, applying pesticides and chemical fertilizers, greenhouse gas emissions and activities related to extracting mines [3]. Cadmium, Lead, Arsenic and Nickel and Zinc to a less extent are all among worrisome factors in fertilizers and modifiers of soil [4]. Fertilizer has been one of the polluting sources of heavy metals, which is harmful for human health and safety of environment [5]. In each sample of agricultural fertilizers (natural fertilizer), concentrations of Cd and Pb did not exceed 0.4 and 4.3 mg kg^{-1} dry weight [6]. Presence of Cadmium in phosphate rock causes contamination of fertilizers with phosphat, so these fertilizers are considered as sources of anthropogenic input in agricultural systems [7]. In a study conducted in south of Iran, the average groundwater Cd concentration level was 0.003 mg L^{-1} and in 4.8% of the samples the level was more than the level of Environmental Protection Agency (EPA) suggested

Corresponding Author: Hajar Boudaghi Malidareh, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran.

Maximum concentration amount of 0.005 mg L^{-1} [8]. Researchers stated that a risk of contamination in the food chain by heavy metals as Arsenic, Lead and Cadmium whose consumption through these agriculture products could cause serious public health problems [9]. Spatial distribution of Pb and Cd showed that the highest level of concentrations had been in northwest of China and gathering of these metals may be the caused by industrial activities, using chemical and agricultural materials and other human operations [10]. Entering of elements coming out of agricultural chemicals to which groundwater was substantial. Also too much use of pesticides and fertilizers is the main reasons behind soil contamination. Average concentration of lead and cadmium in the soil samples was 14 and 0.4 mg kg^{-1}, respectively [11]. The total amount of arsenic in paddy soils of Bangladesh is between 0.3-48.8 mg kg^{-1} [12]. Average concentration of Pb and Cd in paddy soil were 55.8 and 0.66 mg kg^{-1}. Level of concentration of cadmium of soil was more than standard values andirrigation with river water was the main reason for high accumulations of heavy metal in paddy soils [13]. Results of a study in a province called Zhejong (Wenling in Zhejong province) indicated that in some areas under study the potential of contamination of heavy metals was high, especially, Cadmium [10]. The results showed that Mean cadmium concentration in paddy soils in agricultural lands of Iran (Isfahan, Fars and Khuzestan provinces) was 0.45 mg kg^{-1}. Cd concentration in groundwater of southern areas of Iran (Khuzestan Province) was less than amount suggested by WHO and no significant correlation was observed between heavy metal concentration in fertilizer and in consuming fertilizer [14]. Arsenic, Lead and Cadmium concentration in potassium sulfate was 0.24, 4.28 and 0.04 mg kg^{-1}, respectively and their concentration in Triple Super Phosphate type fertilizer was 0.24, 5 and 6.74 mg kg^{-1}, respectively [3]. Rice is a dominant agricultural product in northern parts of Iran. The diseases known as Itai-itai and Minamata were reported due to contamination of paddy rice with Cd and Hg in Japan [15]. Thus, the quality of rice, affects human health greatly. Fertilizers contain heavy metals that can cause serious problems in water and soil. Contents of heavy metals in water and soil are very important because water and soil are the critical first link in the food chain. The purpose of this study was to survey the concentrations of cadmium, lead and arsenic in paddy soil and underground water and its relationship with chemical fertilizers in agricultural fields (paddy soil).

MATERIALS AND METHODS

Ghaemshahr is one of the active zones of agriculture for rice production in Mazandaran province in Northern Iran. Rice cultivation in this city (surrounded area) is 15,650 ha per year. The soil texture of region varies from sandy clay loam to loam. Annual average precipitation in this region is 598 mm for the agricultural period. Vahdat Center is one of the most important areas producing different types of High-yield rice. Rice growing season starts at May and ends up on August or September. Application of N-fertilizer is necessary for rice fields in two periods: one of them before sowing and the other in the middle of the growth stage (Three stage), but generally phosphate fertilizer and potash fertilizer is used before sowing. Agricultural land (in this city) is divided to two regions (The Mountain areas-The plain areas) and five agricultural centers (as shown in due to the rice planting. Considering the high-yielding rice cultivation, expected number of soil samples, status of water supply sources in farming region, the water wells were identified and numbers were given to them randomly. After determination of the number of required samples andthe location of water sampling, paddy land near the well water source,soil sampling was done. Then, 20 composite samples were taken from the soils (paddy soil0-30 cm depth) and groundwater before and after fertilization, before application of fertilizers top-dressing of third. Soil texture was determined by hydrometer method. Soil samples were sieved (<2 mm) and water samples were acidified with HNO_3 (pH≤2). Mean time, composite samples of the three most consumed fertilizers were prepared. Heavy metals (As, Cd and Pb) were extracted by acid digestion method using in soil [HNO3, HCl and H2O2] in fertilizer samples (HNO3) and water samples were prepared after clearing with filter paper, Whatman No. 42.Heavy metals (As, Cd and Pb) in soil, water and fertilizer extracts were measured by inductively coupled plasma-optical emission spectrometer (ICP-OES mod, Germany) [After devices` being calibrated with calibration curve]. The length used for measuring heavy metals has been (As: 189.042 nm), (Cd: 214.438 nm) and (Pb: 220.343 nm) with ICP [16-19].

Statistical analysis comparison of the heavy metal content before and after fertilization (before application fertilizer top-dressingof third) was done by SPSS software ver 11.5.

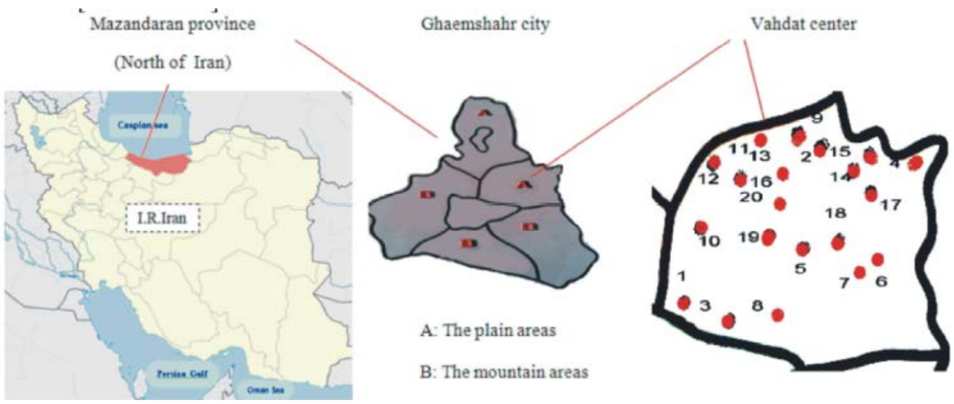

Fig. 1: The area map

Table 1: Average concentration changes of As, Cd and Pb in soil with fertilizer consumption

Average consumed fertilizer (kg ha^{-1})			Change in concentrations (mg kg^{-1})		
Urea	Phosphat	Potash	As	Cd	Pb
272	110	63	0.222	0.049	0.317

Table 2: Heavy metal concentrations in three fertilizer of high

Applied fertilizers	As	Cd	Pb
Urea	0.020	0.000	0.010
Triple super	5.230	2.980	1.470
Phosphate potassium sulfate	0.187	0.015	1.047

RESULTS

Fertilizer

Consumed Fertilizer: The local and high-yield cultivation of rice in the study area is 20 and 80%, respectievly. The percentage of rice cultivation with high-yielding rate (n = 20) to the total (4003 ha) area under cultivation in the area)Vahdat Center(was equal to 1.02%. Total fertilizer consumption of urea, phosphate and potashin studied area for high-yielding rice varieties were 8700, 3085, 1355 (kg) or 272, 110, 63 kg ha^{-1} and Average concentration changes of As, Cd and Pb in soil with fertilizer consumption were As (0.222), Cd(0.049) and Pb(0.317) mg kg^{-1} respectively, Table 1.

Heavy Metal concentrations In Fertilizers: The order of the concentrations of As, Cd and Pb in three different analyzed fertilizers according to the Table 2 are as follow: triple super phosphate>sulfate, potassium>urea. The cadmium concentration in urea type was lower than detection limit.

Water: The results of concentrations of As, Cd and Pb in well water samples with depth 11-70 m, have shown that in both sampling stages the concentrations of all three metals was zero. pH of water wells at the time of sampling was in the range of 7.1-7.4.

Paddy Soil: Average arsenic concentrations of soil samples before fertilization and after fertilization (Before application fertilizer top-dressing of third) were 0.003 (0.001-0.007) and 0.225 (0.1-0.3) mg kg^{-1}, respectively. According to the analysis of soil in the area, the average electrical conductivity of soil samples were 1.2 μm cm^{-1} (0.43-1.78) and the average pH in soil was 7.8 (7.34-8.5). The results showed that the average soil cadmium concentration before and after fertilization (Before application fertilizer top-dressing of third) were zero and 0.049 (0.045-0.052 mg kg^{-1}), respectively. Also, the average

Table 3: Comparison of soil Heavy metal concentrations in soil paddy in mg kg^{-1} (before and after fertilization)

	Arsenic			Lead			Cadmium		
	Before	After	Changes	Before	After	Changes	Before	After	Changes
Mean	0.003	0.225	0.222	0.089	0.311	0.222	0.000	0.049	0.049
SD	0.002	0.044	0.044	0.012	0.062	0.060	0.000	0.002	0.002
Min	0.001	0.100	0.096	0.066	0.201	0.102	0.000	0.045	0.045
Max	0.007	0.300	0.298	0.103	0.447	0.344	0.000	0.052	0.052

Table 4: Amount of metal added (g/ha/y) to the paddy soil and water and rice system in the process of fertilization

Average consumed Fertilizer (kg ha^{-1})			Amount of metal added to soil and water or rice(g/ha/y)		
Urea	Phosphat	Potash	As	Cd	Pb
272	110	63	0.179	0.578	0.398

Note: Each composite fertilizer sample include 5 specimens

lead concentrations in soil samples before fertilization were 0.089 (0.066-0.103 mg kg^{-1}) and after fertilization were 0.311 (0.201-0.447 mg kg^{-1}), Table 3.

DISCUSSION

There is no available information on typical background values for Iranian soils. Average concentration of Lead and Cadmium was lower than their mean value in industrial areas in Hamadan Province [20]. Researchers stated that with increasing the requisition periods from 1 to 3 years, Cadmium and Lead concentration increased in soil [21]. According to preventing rule of Japan, Arsenic concentration was lower than the concentration in agricultural soil. About 15 mg kg^{-1} [22]. Amount of consumption of urea and phosphate fertilizer was 1.1 and 2.2 times more than fertilizer recommendations, respectively. Generally, minimum changes of arsenic and cadmium concentration in soil (before and after fertilization) were less than lead concentration changes. Manure, sludge and atmospheric deposition are the most important sources of Lead in agricultural soils [23]. Arsenic is widely found in nature. It is followed by igneous and sedimentary rocks, especially with sulphide ores. Following is the factors which are responsible for the emission of arsenic into the atmosphere natural phenomena: biological activity, volcanic activity, weather condition along with anthropogenic inputs. Soil levels of the Ganges river flood plain were higher compared to those of Meghna river floodplain. Right now Bangladesh is facing challenge of high level of arsenic concentration in shallow aquifers. Total amount of Arsenic in soils was correlated with clay content, soil pH and Fe oxide positively. The results showed that fine textured, high pH soils have higher Arsenic content to coarse textured soils [12]. The results showed that arsenic content in soil ranged between 0.25 and 255 mg kg^{-1} and other place between 5.5 and 295 mg kg^{-1} [24]. Observations conducted in deferent parts of Bangladesh also indicated that iron-arsenic and phosphorus-arsenic ratios in groundwater may vary a lot and this variation may be significant for as accumulation rates in soils, plant uptake and mitigation measures [25]. The concentration of Arsenic in water ranged between 0.00 – 0.37 ppm. The result showed that, in seven water samples have low level of Arsenic metal pollution with values below 0.05 ppm WHO standard and that Arsenic pollution result from human activities [26]. The mean Pb concentration of groundwater was 0.25±0.16 (wet season), greater than WHO standard of 0.01 mg l^{-1} [27]. Comparing rate of heavy metals in the area studied (Kermanshah of Iran) with some other studies in various countries, we had the following results: rate of as (0.179 g/ha/y) in this study was lower than the rates in the Netherlands (3.2g/ha/y), middle Europe (2 g/ha/y) and Denmark (1.1 g/ha/y). The rate of cadmium (0.578 g/ha/y) was lower than the rates in the Netherlands (1.3 g/ha/y), Europe average (1.9 g/ha/y) as well, but it was more than the rates in Denmark (0.3 g/ha/y) and Finland (0.2 g/ha/y) to some extent. Level of lead (0.398 g/ha/y) was lower than all favorable countries,Table 4. Level of arsenic concentration in Urea, Triple Super Phosphate and Potassium Sulfate is 0.02, 1.47 and 0.187 mg kg^{-1}, respectively. Also, Cd concentration in desired fertilizers was 0.00, 5.23 and 0.015 mg kg^{-1} and for lead it was 0.001, 2.98 and 1.047 mg kg^{-1}, respectively. Heavy metals were lower than the levels that could be useful for agricultural products (Atafar *et al*., 2010). Theresults showed that heavy metal concentrations are lower than world standards. Cadmium concentration in Triple Super Phosphate and Urea fertilizers is lower than cadmiumwhich is used by chemical fertilizers in Shush and Andimeshk aquifer (Triple Super Phosphate = 12.2 mg kg^{-1} and Urea = 0.03) (Nouri *et al*., 2008). The only higher case in the results is that Cd concentration in Triple Super Phosphate fertilizer is higher than CDFA standard The only significant relationship between the amount of potash fertilizer and cadmium concentration in soil ($p<0.05$) was observed. The highest concentration of Cd in phosphate fertilizeris due to the lands that used more potash fertilizer than usuall consumption. But, according to the presence of possitive relationship between the phosphate fertilizer and potash fertilizer ($p<0.05$), the reason is not really known. Heavy metal concentrations in underground water in agricultural area was zero. No relationship between the amount of heavy metals in the soil with water and heavy metals in the fertilizer with water was observed, Table 5-7 and Fig. 2.

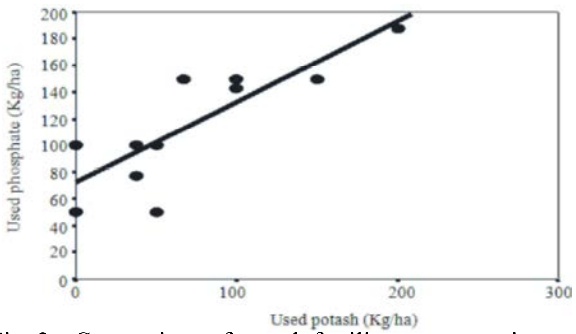

Fig. 2: Comparison of potash fertilizer consumption and phosphate fertilizer in soil

Table 5: Relationship between change in concentration of As, Pb and Cd in soil and consumed fertilizer(P-value)

Compare the fertilizer with metal	Significant	Compare the fertilizer with metal	Significant
Urea and As	0.944	Potash and Pb	0.213
Urea and Cd	0.703	Phosphat and As	0.726
Urea and Pb	0.763	Phosphat and Cd	0.097
Potash and As	0.852	Phosphat and Pb	0.610
Potash and Cd	0.041		

Table 6: Relationship between fertilizer of urea with potash fertilizer

	Correlations	Urea	Potash
Urea	Pearson correlation	1.000	-0.172
	Sig. (2-tailed)	0.000	0.469
	N	20.000	20.000
Potash	Pearson Correlation	0.172	1.000
	Sig. (2-tailed)	0.469	0.000
	N	20.000	20.000

Table 7: Relationship between fertilizer of phosfat with potash fertilizer

	Correlations	Potash	Phasphate
Potash	Pearson Correlation	1.000	0.765(**)
	Sig.(2-tailed)	-0.000	0.000
	N	20.000	20.000
Phasphate	Pearson Correlation	0.765(**)	1.000
	Sig.(2-tailed)	0.000	-0.000
	N	20.000	20.000

CONCLUSION

Although concentration of heavy metals in soil and water has not exceeded before and after fertilization in this study, but frequent use of chemical fertilizers and pesticides in common agricultural fields can cause increase in rate of heavy metals in soil and water. Also, considering the results, it's apparent that concentration of all heavy metals (As, Cd, Pb) has increased during the crop year. In this study after fertilization sampling was conducted before pesticides loads and minimum use of it that effect of chemical pesticides on concentration of heavy metals in the soil and water is considerable. In more cases, it was observed that the consumed fertilizers were without the proper production and expiration dates. So, because of reduction of products effectiveness, farmers had to use higher amount of the fertilizers then officialy recommended which results in environmental pollution. According to management strategies, reduction of environmental side effects of fertilizers is recommended. Testing the soils in agricultural lands to determine the real needs of paddy lands to fertilizers, distribution of allowed and standard fertilizers through governmental agencies set national and provincial standards, use of fertilizers compatible to environment, monitoring land owners on farmer's education and performance is needed. According to the results gained, annual values of lead, As and Cd added to soil are 1880, 2980 and 200 times, respectively which are more than value of heavy metals which are added to paddy soil by consuming fertilizer. On the other hand, value of heavy metals gained in the soil is low compared to the results gained in the soil of countries such as Bangladesh or Iran in which we believe several results gained can be involved:

- Using traditional agriculture
- Characteristics of consuming water for irrigation
- Producing product just in the amount needed, as a result lack of need to more production with new methods or with less fertilizer consumption
- Changing application of lands in sequent years (residential, green farms, fruit gardens, paddy land with producing local rice, high-yield rice)
- Exist of other resources of heavy metals in soil (chemical poisons and so on). It is worth mentioning that for the least effect of chemical poisons on the results, the second sampling was conducted before application of fertilizers top-dressing of third and using chemical poisons
- Attracting some values of heavy metals annually and by producing products (in the lands after harvesting rice, other products are also planted such as wheat, vegetable)
- Also its expected health authorities study annual level of heavy metals in soil and groundwater.

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