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HEAVY METAL CONCENTRATIONS IN IRRIGATION WATERS AND RICE CULTURE IN THE CENTRAL TRAKYA REGION

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Abstract. In this survey, irrigation water, leaves, root, rice grains and husk samples were taken during the vegetation period of the rice plant from 50 different rice fields of Meric and Uzunkopru provinces in the Central Trakya region. In the samples Fe, Zn, Cu, Mn, Cd, Ni and Pb contents were determined by AAS and various correlations were calculated. Results showed Fe and Mn concentrations to be at toxic levels at all plant parts studied. Zn, Pb and Ni levels were also found to be at toxic concentrations in the plant roots. Additionally, Cu was also at toxic concentrations in both husk and roots. The heavy metal contents of the plant tissues, polluted by the irrigation water of the Ergene river were determined to be as follows: root > leaf > grain > husk. Heavy metal pollution in the rice fields, originating from the use of irrigation water from the Ergene river, was determined to be concentrated especially in the roots. The plant tissues contained increasing levels of heavy metals as the heavy metal content of the irrigation water increased. Overall, the research showed that the heavy metal pollution is on the increase in the ecological circle concerned.

Keywords: heavy metals, irrigation waters, rice culture.

AIMS AND BACKGROUND

The Ergene river has become increasingly polluted in recent years due to industrial and domestic wastes especially originating from the ever-expanding industry of the Central Trakya region, Turkey. Generally, waters of the Ergene river are used in the irrigation of rice crops in the Uzunkopru and Meric area of the Edirne province where the research presented here was carried out. In the crops such as rice, sunflower and wheat irrigated with water from the Ergene river, probability of heavy metal pollution creates concern. The case is even worse for the rice grown in soils saturated with such polluted waters. The heavy metals accumulated in the rice plants and grains threaten health both of humans and animals.

In the cited in Ref. 1 study on heavy metal contents of aquatic plants and sediments in lakes and four rivers in the Macedonia, Greece, heavy metal accumulation in the species studied varied depending on the lakes or rivers studied. Lower levels of heavy metals were determined in the lakes in comparison to the

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rivers studied. Also in the study it was determined that average heavy metal contents of the sediments and aquatic plants were as follows: Mn > Zn > Ni > Cu > Pb > Cd. Degree of heavy metal accumulation in the plant organs was as follows: roots > tubers > leaves > flowers > stem > seed.

In a separate research[2] carried out to determine the level of heavy metal pollution in the city of Zhuzhou it was determined that heavy metals accumulated, with a very slow horizontal movement, in the top layers of the soils. Also in this study, heavy metal concentrations in the vegetables and rice were above the toxicity thresholds.

The accumulation of heavy metals in soils and plants irrigated with polluted water with heavy metals was investigated also in China. Results showed that the areas irrigated with such polluted waters contained high levels of Cd and Zn. Levels of the heavy metals accumulated in plants varied according to the plant species and its capability of absorption of the metals. Consumption of rice grown in rice fields contaminated with Cd, Cr and Zn posed a serious threat to human health, the reason for which is, according to the plant characteristics of rice, 22-24% of the heavy metals absorbed accumulate in the rice grains[3].

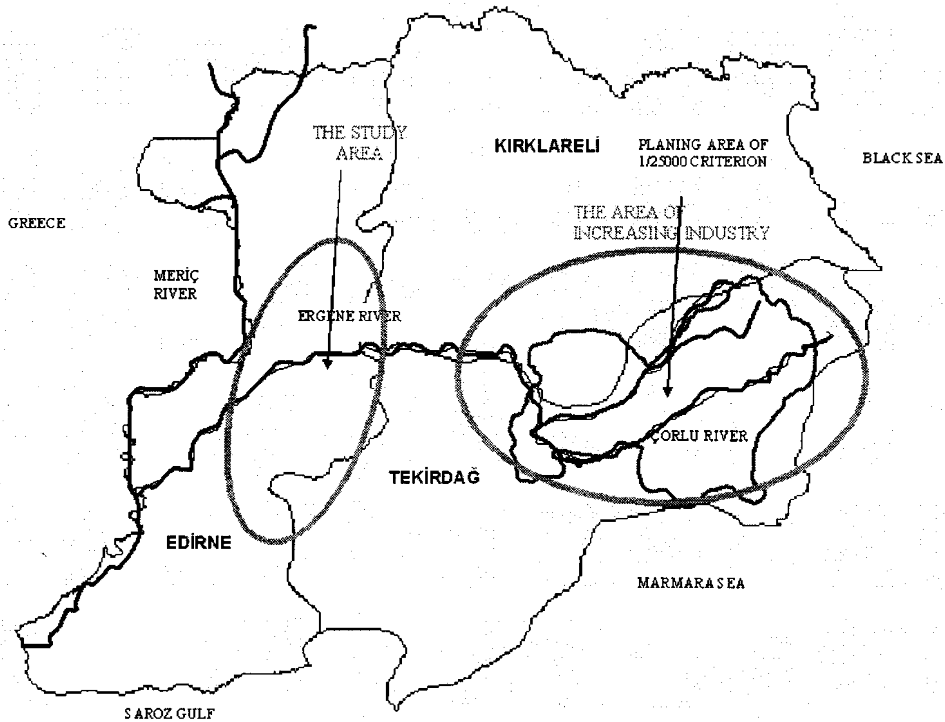
The aims of this study were to determine heavy metal contents of the irrigation water and various parts of the rice plants and finally to find out a possible relation between the heavy metal concentrations of the water and the rice plants.

EXPERIMENTAL

A total of 50 samples, each from a separate rice field, were collected from the Meric and Uzunkopru areas, where rice cultivation is practiced intensively, and the sampling continued throughout the vegetation period with 15 days intervals. The first, second, third and fourth dates of sampling were 26.07.2001, 02.08.2001, 16.08.2001 and 06.09.2001, respectively. During sampling, water, leaf and root samples were taken. Finally, on 10.10.2001, at the harvest time, grain and husk samples were also taken. Studied area is given in the following map.

All the samples were immediately brought to the laboratory and the pH of the water samples was reduced to 2.0 for processing later[4]. The leaf and root samples on the other hand following washing with distilled water were air dried and after grinding were ready for analyses. The samples of grain were separated into the grain and husk and after grinding were ready for digestion.

Plant samples of 1.0 g were topped with 12 ml of a 1: 3 mixture of nitric-perchloric acid and wet acid digested samples were placed immediately on hot plate at 180-200°C. The digested samples were then filtered through the white striped filter papers (Whatman No 42) by washing with distilled water. The precipitated material was then collected in volumetric flasks and so prepared



Location of studied area in Trakya region

samples were analysed with atomic absorption spectrophotometer[5,6]. After the filtration, the water samples were also analysed by the same equipment[5].

In order to check the recoverability of the results obtained from the AAS, a total of 40 samples, selected randomly, were analysed using the inductively coupled plasma (ICP) and subsequently correlations between the results of ICP and AAS were determined. Significant positive correlations were obtained for all of the elements following statistical analyses.

RESULTS AND DISCUSSION

IRRIGATION WATERS

In the water samples the heavy metal concentrations were as follows: iron concentrations, for all four sampling dates, varied between 0.10 and 17.80 ppm. The average Fe contents for the 1st, 2nd, 3rd and 4th sampling dates were 0.54, 0.71, 1.53 and 1.91 ppm, respectively (Fig. 1). In the samples of all the sampling dates, Fe concentrations of the waters tested fell into the class IV of the SKKY (Water Pollution Control Regulation, Turkey), issued on 4 September 1988 in the State newspaper with the article No 19919.

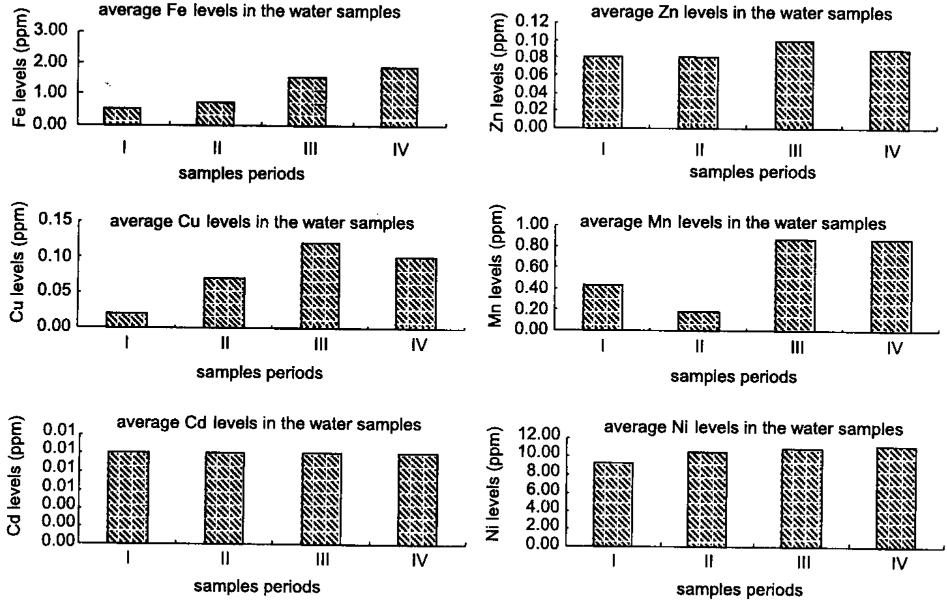


Fig. 1. Average element contents of water samples

In all the sampling dates, regarding the Zn concentrations the minimum and maximum levels ranged between 0.02 and 0.19 ppm, respectively, whereas the averages for all the sampling dates, in the progressing order, were 0.07, 0.08, 0.10 and 0.09 ppm, respectively (Fig. 1). In the samples of all the sampling dates, Zn concentrations of the waters tested fell into the class I of the SKKY.

Copper concentrations, on the other hand, in the water samples tested varied between 0.02 and 0.11 ppm in all four sampling dates. For the 1st, 2nd, 3rd and 4th sampling dates the average values were 0.02, 0.07, 0.11 and 0.10 ppm, respectively (Fig. 1). The SKKY classes of the Cu concentrations were II, III, IV and IV in the sampling dates of the progressing order.

The minimum and maximum Mn concentrations in the samples in all the sampling dates were 0.01 ppm and 5.20 ppm, respectively. For the 1st, 2nd, 3rd and 4th sampling dates the average Mn concentrations were 0.42, 0.18, 0.87 and 0.88 ppm, respectively (Fig. 1). The Mn concentrations were in the class II of the SKKY for all the sampling dates.

In all the sampling dates, regarding the Cd concentrations the minimum and maximum levels ranged between 0.00 ppm and 0.03 ppm, respectively. For the 1st, 2nd, 3rd and 4th sampling dates the average Cd concentrations were 0.01, 0.01, 0.01 and 0.01 ppm, respectively (Fig. 1). The SKKY class of the Cd concentrations for all the sampling dates was I.

The concentrations of the last element tested, i.e., Pb, in the water samples varied between 0.00 and 45.49 ppm in all four sampling dates. The average Pb concentrations in all the sampling dates of progressing order were 9.21, 10.44, 10.87 and 11.21 ppm, respectively (Fig. 1). According to the SKKY the Pb concentrations determined in all the sampling dates were in the class IV.

The toxicity limits of elements in water were determined earlier and according to Ref. 7 the concentrations in water of Cd, Cu, Pb, Ni and Zn up the levels of 0.005, 0.2, 5.0, 0.5 and 5.0 ppm, respectively (Fig. 1), are tolerable and above these levels are toxic. Also, according to the DSI (State Water Authority of Turkey, 1976) the levels of Fe, Cu and Mn in the irrigation water above 2.0, 0.2 and 2.0 ppm, respectively, are toxic (Table 1).

Table 1. Toxicity limits for irrigation waters

Elements	Toxicity limits (ppm)	References
Fe	2.0	DSI, 1976, Ref. 8
Zn	5.0	Ref. 7
Cu	0.2	Ref. 7 and DSI, 1976
Mn	2.0	DSI, 1976
Cd	0.005	Ref. 7
Pb	5.0	Ref. 7
Ni	0.5	Ref. 7

Our results for the water samples are shown in Fig. 1 which when evaluated according to the toxicity limits of Table 1 the following conclusions can be stated: the concentrations of Fe were low in general whereas in the samples of 3rd and 4th sampling date an increase exceeding the toxic level of 2 ppm was observed.

In all the water samples, Zn levels determined were below the toxic concentration of 5 ppm (Fig. 1).

Except for few samples of water, the Cu concentrations were determined to be under toxic levels. The concentrations of Cu in the samples of the 1st sampling date, in comparison to the others, were at low levels, which gradually increased as the sampling dates progressed.

According to the analyses of the water samples Mn concentrations measured were below the toxic level in general and the Mn concentrations varied from one field to another during the vegetation period. However, an increase towards the end of the vegetation period was observed.

In general, concentrations of Cd and Pb in the water samples were above the toxic levels of 0.005 and 5.0 ppm, respectively.

The toxicity limits of the heavy metals in plants according to Ref. 9 in rice (*Oryza sativa* L.) are as follows: Zn (30-70 ppm); Cu (7-12 ppm); Mn (40-100 ppm) whereas according to Ref. 10 the toxic Fe concentrations in plants varies between 50-150 ppm. For the authors in Ref. 11 non-toxic levels in the plants of Cd, Pb and Ni are 0.05-0.2, 5-10 and 0.5-5 ppm, respectively (Table 2). According to the above mentioned limits the Fe and Mn levels, determined in the present study, were in toxic concentrations in all the plant parts tested. While the Zn, Pb and Ni were in toxic concentrations in the roots, the Cu was determined to be over the toxic levels both in husks and roots (Table 2).

Table 2. Heavy metal concentrations of plants in normal levels

Elements	Toxicity limits (ppm)	References
Fe	50-150	Ref. 10
Zn	30-70	Ref. 9
Cu	7-12	Ref. 9
Mn	40-100	Ref. 9
Cd	0.05-0.2	Ref. 11
Pb	5-10	Ref. 11
Ni	0.5-5	Ref. 11

Leaf. In the leaf samples the heavy metal concentrations were as follows: iron concentrations, for all four sampling dates, varied between 15.70 and 1590.70 ppm. The average Fe contents for the 1st, 2nd, 3rd and 4th sampling dates were 202.81, 372.09, 402.84 and 358.63 ppm, respectively (Fig. 2). The above mentioned Fe concentrations determined in the leaf samples were over the toxic limit in general which is possibly due to the excess Fe content of the irrigation water.

In all the sampling dates, regarding the Zn concentrations the minimum and maximum levels ranged between 0.10 and 58.30 ppm, respectively, whereas the averages for all the sampling dates, in the progressing order, were 27.10, 29.17, 31.74 and 22.76 ppm, respectively (Fig. 2). Therefore, Zn contents of the leaf samples were below the toxic levels.

Copper concentrations, on the other hand, in the leaf samples tested varied between 0.50 and 16.10 ppm in all four sampling dates. For the 1st, 2nd, 3rd and 4th sampling dates the average values were 6.26, 5.51, 7.36 and 7.89 ppm, respectively (Fig. 2). Except for a few samples Cu concentrations were below the toxic level.

The minimum and maximum Mn concentrations in the samples in all the sampling dates were 54.20 and 935.70 ppm, respectively. For the 1st, 2nd, 3rd and 4th sampling dates the average Mn concentrations were 346.21, 395.86, 358.37

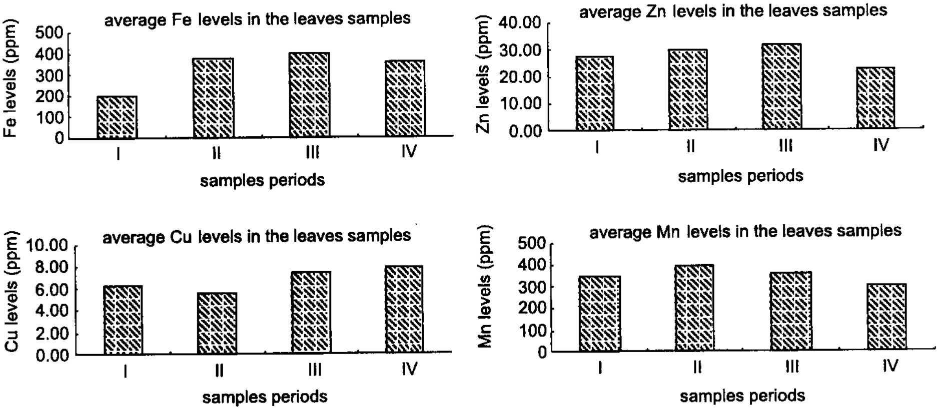


Fig. 2. Average element contents of leaf samples

and 295.66 ppm, respectively (Fig. 2). In all the samples Mn levels were above the toxic limit which in some of the samples reached upto eight times that of the toxic limit.

Roots. In the root samples the heavy metal concentrations were as follows: iron concentrations, for the last two sampling dates, varied between 608.70 and 26418.70 ppm. The average Fe contents for the 3rd and 4th sampling dates were 9993.70 and 8781.02 ppm, respectively (Fig. 3). Fe contents of the roots were much higher than the toxic limit of 150 ppm. The high Fe concentrations determined in the

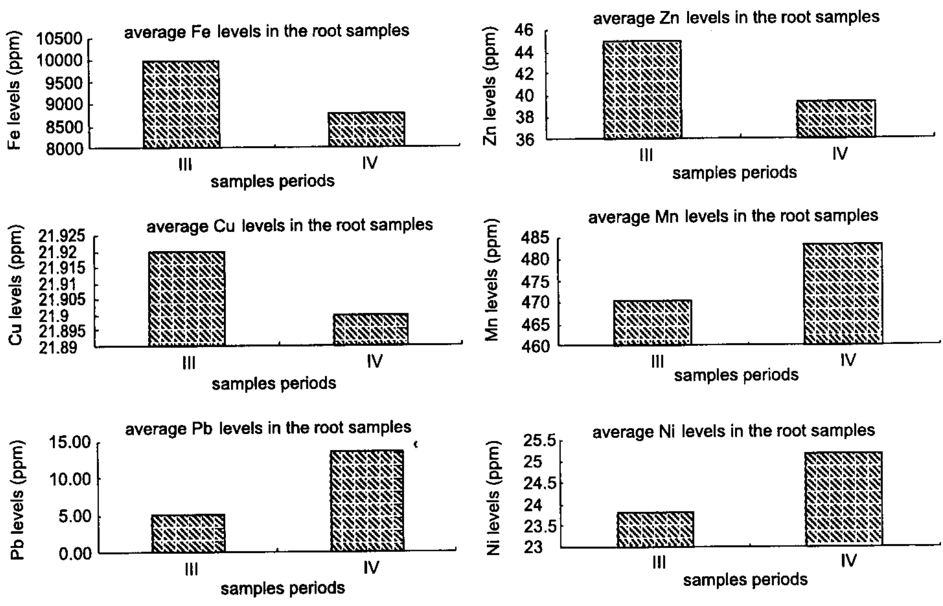


Fig. 3. Average element contents of root samples

roots were parallel to the increase of the Fe concentrations in the irrigation waters which increased with the progressing dates of sampling.

In the 3rd and 4th sampling dates, regarding the Zn concentrations the minimum and maximum level ranged between 16.20 and 88.50 ppm, respectively, whereas the averages for the same sampling dates were 44.91 and 39.42 ppm, respectively (Fig. 3). Except for some of the root samples, Zn toxicity was not determined in the roots.

Copper concentrations, on the other hand, in the root samples tested varied between 1.70 and 51.20 ppm in the last two sampling dates. For the 3rd and 4th sampling dates the average values were 21.92 and 21.90 ppm, respectively (Fig. 3). In general, Cu contents of the root samples were above the toxic levels.

The minimum and maximum Mn concentrations in the samples of the 3rd and 4th sampling dates were 150.00 and 917.00 ppm, respectively. For the 3rd and 4th sampling dates the average Mn concentrations were 470.41 and 483.41 ppm, respectively (Fig. 3). Mn contents of the root samples were above the toxic levels, which in some cases reached eight times that of the toxic limit. The authors cited in Ref. 1, as mentioned above, obtained similar results in the case of roots. The authors in Ref. 12 indicate also that under waterlogged conditions reducing processes dominate and thus provide a high level of Mn availability which may even result in Mn toxicity.

In the 3rd and 4th sampling dates, regarding the Pb concentrations the minimum and maximum levels ranged between 0.00 and 53.65 ppm, respectively. For the 3rd and 4th sampling dates the average Pb concentrations were 5.13 and 13.64 ppm, respectively (Fig. 3). The toxic levels of Pb determined in the root samples were possibly due to the fact that the mobility of Pb is of a slow type hindering its movement to the upper parts of the plant[13].

The concentrations of the last element tested, i.e., Ni, in the root samples varied between 1.27 and 91.09 ppm in the 3rd and 4th sampling dates. The average Ni concentrations in the 3rd and 4th sampling dates were 23.82 and 25.20 ppm, respectively (Fig. 3). It was determined that the Ni, in general, accumulated at toxic levels in the roots.

In general it can be concluded that heavy metal accumulation at toxic levels was determined in the root samples and the heavy metal concentrations determined were higher in the 3rd sampling date in comparison to the 4th one.

Grain. In the grain samples the heavy metal concentrations were as follows: the minimum and maximum Fe concentrations in the grain samples were 39.66 and 918.68 ppm, respectively, whereas the average Fe concentration was 156.54 ppm (Fig. 4).

Zinc concentrations in the grain samples varied between 11.98 and 46.44 ppm and the average concentration was 32.04 ppm (Fig. 4).

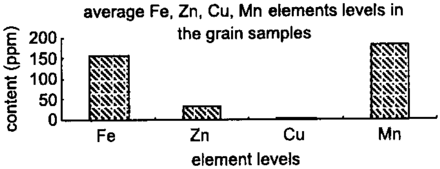


Fig. 4. Average element contents of grain samples

Copper concentrations in the grain samples were determined to be at minimum 0.00 ppm and maximum 7.42 ppm whereas the average concentration was 2.66 ppm (Fig. 4).

Regarding the Mn concentrations in the grain samples, the minimum and maximum concentrations determined were 62.88 and 340.37 ppm whereas the average value was 182.49 ppm (Fig. 4).

Husk. In the husk samples the heavy metal concentrations were as follows: the minimum and maximum Fe concentrations in the grain samples were 20.10 and 1074.70 ppm, respectively, whereas the average Fe concentration was 103.10 ppm (Fig. 5).

Zinc concentrations in the husk samples varied between 19.20 and 47.80 ppm and the average concentration was 30.40 ppm (Fig. 5).

Copper concentrations in the husk samples were determined to be at minimum 1.90 ppm and maximum 39.70 ppm, whereas the average concentration was 14.39 ppm (Fig. 5).

Regarding the Mn concentrations in the husk samples, the minimum and maximum concentrations determined were 0.00 and 417.50 ppm, respectively, whereas the average value was 34.82 ppm (Fig. 5).

In general, it can be stated that Fe accumulated at a higher level in the grains in comparison to the husks. Despite the fact that the situation was the opposite in 10 of the samples tested, it was determined that the Fe accumulated at a higher level in the husks. The analyses showed that Zn was not at the toxic levels in grain and husk samples. Cu accumulation, on the other hand, was at a toxic level in the husks, whereas the level of accumulation was below the toxic level in the grains. Manganese accumulation, however, was at a toxic level in the grains, whereas the level of accumulation was below the toxic level in the husks.

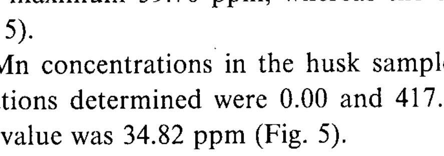


Fig. 5. Average element contents of husk samples

Table 3. Concentrations of heavy metals in the water, leaf, root, grain and husk samples

	Sample periods		Fe	Zn	Cu	Mn	Cd	Pb	Ni
Water	I	interval of change average	0.24-2.04 0.54	0.05-0.11 0.08	0.00-0.06 0.02	0.05-5.20 0.42	0.00-0.03 0.01	0.00-45.49 9.21	trace
	II	interval of change average	0.12-2.92 0.71	0.04-0.13 0.08	0.02-0.12 0.07	0.03-0.82 0.18	0.00-0.02 0.01	0.00-44.64 10.44	trace
	III	interval of change average	0.10-17.80 1.53	0.02-0.19 0.10	0.05-0.37 0.12	0.04-9.40 0.87	0.00-0.02 0.01	0.00-33.83 10.87	trace
	IV	interval of change average	0.16-10.80 1.91	0.05-0.17 0.09	0.05-0.20 0.10	0.01-4.85 0.88	0.00-0.03 0.01	0-23.86 11.21	trace
Leaves	I	interval of change average	28.58-1100.70 202.81	0.10-58.30 27.10	0.50-12.20 6.26	127.20-671.90 346.21	trace	trace	trace
	II	interval of change average	15.70-1590.70 372.09	16.60-45.30 29.17	1.60-16.10 5.51	92.00-935.70 395.86	trace	trace	trace
	III	interval of change average	39.40-1178.70 402.84	6.00-58.20 31.74	1.60-15.90 7.36	54.20-810.20 358.37	trace	trace	trace
	IV	interval of change average	99.30-892.70 358.63	9.00-35.10 22.76	2.10-14.80 7.89	85.00-690.90 295.66	trace	trace	trace
Root	III	interval of change average	608.70-26418.70 9993.70	20.40-88.50 44.91	10.00-51.20 21.92	180.40-917.00 470.41	trace	0.00-37.33 5.13	3.08-91.09 23.82
	IV	interval of change average	2823.70-20388.70 8781.02	16.20-76.80 39.42	1.70-45.20 21.90	150.00-916.30 483.41	trace	0.00-53.65 13.64	1.27-74.86 25.20
Grain		interval of change average	39.66-918.68 156.54	11.98-46.44 32.04	0.00-7.42 2.66	62.88-340.37 182.49	trace	trace	trace
Husk		interval of change average	20.10-1074.70 103.09	19.20-47.80 30.40	1.90-39.70 14.39	0.00-417.50 34.82	trace	trace	trace

I – taken samples at 26.07.2001; II – taken samples at 02.08.2001; III – taken samples at 16.08.2001; IV – taken samples at 06.09.2001.

Our results showed that heavy metals accumulated in the parts of the rice plants in the following order of increase: root> leaf> grain> husk. In the grains, the elements of Fe and Mn were over the toxic levels, which poses the threat both to the human and animal health. As indicated by the authors cited in Ref. 14, the distribution of the heavy metals in the organs of the studied crops was also revealed in the same order: root>stems>fibre>leaves>seed.

RELATIONSHIPS OF HEAVY METAL CONTAMINATION BETWEEN WATER, PLANTS AND SAMPLING

In the statement below of the relationships of heavy metal contamination between water, plants and sampling dates (Table 3) only statistically significant correlations are presented. Statistical analyses carried out for the Fe concentrations in the leaf samples significant positive correlations were determined between the 1st and 2nd sampling dates at 5% level; between 4th sampling date of the leaf samples and the same sampling date of the water samples at 1% level; and between grain and the husk samples at 1% level (Table 4).

Table 4. Correlation coefficients for the Fe between the samples of water and plant

	Fe leaf-II	Fe husk	Fe water-IV
Fe leaf-I	0.347*		
Fe leaf-IV			0.357**
Fe grain		0.633**	

* Relation at the level of 5 %; ** relation at the level of 1 %.

Statistical analyses carried out for the Zn determined significant correlations at 1% level between the 2nd and the 3rd sampling dates; for the leaf samples between the 3rd and 4th sampling dates at 1% level; between the grain and the husk at 1% level; beteween the 3rd sampling date of the leaf samples and the 3rd sampling date of the roots at 5% level; between the root sampling dates of 3rd and 4th at 5% level. A negative correlation at 1 % level between the 1st and 3rd sampling dates was determined (Table 5).

Table 5. Correlation coefficients for the Zn between the samples of water and plant

	Zn leaf-III	Zn leaf-IV	Zn husk	Zn root-III	Zn root-IV	Zn water-III
Zn leaf-II	0.360**					
Zn leaf-III		0.484**		0.321*		
Zn grain			0.439**			
Zn root-III					0.338*	
Zn water-I						−0.386**

* Relation at the level of 5 %; ** relation at the level of 1 %.

Statistical analyses carried out for the Cu concentrations of the plant and water samples showed a significant correlation at 1% level between 3rd and the 4th sampling dates of the root samples; a significant correlation at 5% level between the second sampling dates of the husk and water; also at 5% level a significant correlation between the water samples of the 3rd sampling date and the leaf samples of the 4th sampling date as determined (Table 6).

Table 6. Correlation coefficients for the Cu between the samples of water and plant

	Cu root-IV	Cu water-II	Cu water-III
Cu leaf-IV			0.287*
Cu husk		0.336*	
Cu root-III	0.365**		

* Relation at the level of 5 %; ** relation at the level of 1 %.

Significant correlations between the plant and water samples for the Mn were as follows: a significant correlation at 1% level between the 3rd and 4th sampling dates of leaf samples; a significant correalation at 1% level between the husk and the water samples of the second sampling dates was determined. The parameters for which significant positive correlations determined at 5% level were as follows: between the 1st sampling date of the leaves and the husks; between 1st sampling date of the leaves and the 2nd sampling date of water; between the 3rd leaf sampling date of the leaf and 4th sampling date of the root; between the 4th sampling date of leaves and the grains; between the 3rd and 4th sampling dates of water; whereas a negative correlation was determined between the 2nd sampling date of the leaves and the 3rd sampling date of the roots (Table 7).

Table 7. Correlation coefficients for the Mn between the samples of water and plant

	Mn leaf-IV	Mn grain	Mn husk	Mn root-III	Mn root-IV	Mn water-II	Mn water-IV
Mn leaf-I			0.283*			0.333*	
Mn leaf-II				−0.293*			
Mn leaf-III	0.391**				0.290*		
Mn leaf-IV		0.291*					
Mn husk						0.570**	
Mn water-III							0.303*

* Relation at the level of 5 %; ** relation at the level of 1 %.

According to statistical analyses for the Pb concentrations, a significant positive correlation at 1% level ($r = 0.434^{**}$) between the water samples taken at the 3rd and 4th sampling dates was determinated.

CONCLUSIONS

In this study Fe and Mn have been observed at toxic levels in all parts of the plant, whereas toxic levels of Zn, Pb and Ni have only been observed in the roots of the rice plant. On the other hand, Cu has been observed above the toxic levels only in both the husk and the roots of the rice plant.

The heavy metal contents of the plant organs, polluted by the irrigation water deviated from the Ergene river were determined to be as follows: root > leaf > grain > husk. Therefore, we can state that the heavy metal pollution in the rice fields was determined to be concentrated, especially in the roots. The plant tissues contained increasing levels of heavy metals as the heavy metal content of the irrigation water increased. Overall, the research showed that the heavy metal pollution is increasing in the ecological circle concerned.

As the level of contamination reached higher levels, rice plants became affected from this contamination directly and this led to a high heavy metal accumilation in the roots of the plants as the contamination eventually spread to other parts of the plant, finally resulting in a high level of heavy metal contamination in the grains of the rice plants.

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