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FATE OF METALLOID FUNGICIDE IN WATER, SOIL AND PLANT IN EGYPT

Heavy metals are among the most troublesome elements in the environment. Their adverse impact on human health is besides several harmful phenomena worldwide. Nowadays we deal with heavy metals in a normal everyday life in industrial products and agro-chemicals, e.g. fertilizers and pesticides. These metals readily find their way to surface waters and, hence, to the food chain. The present study aimed at tracing up the fate of the heavy metal content in water, soil and potato crop if the fungicide Mancopper is used for field and pot experiments. The fungicide was employed in Qatta Village in Giza Governorate, Egypt. The experiments were carried out for over four successive seasons and mean levels of the metals of interest, namely Fe, Zn, Mn, Cd and Pb, were determined. The levels of the metals in the surface layer of soil irrigated with the Nile water and ground water were arranged in the following ascending orders Fe > Zn > Mn > Pb > Cu > Cd and Fe > Mn > Zn = Cu > Pb > Cd, respectively. The parts of plants (potatoes) grown on irrigated soil can be arranged in the following ascending order: root > leaf > stem > tuber if their metal content is taken into account. Irrespective of the virtual least metal content in the edible part (tuber), the levels recorded for copper and lead exceeded their permissible values. Careful pot experiments under controlled conditions were compared with field experiments under conditions of fungicide treatment, particularly in respect of high content of copper. It was also concluded that in spite of its relatively low concentration in the fungicide, lead revealed considerably high concentration in soil and plant parts. This illustrates active patterns of metal accumulation in soil and its uptake by plants. It was eventually recommended that further efforts should be made to cope with the deteriorating situation of such strategic crop by limiting heavy metal application and minimizing their contents in the soil so as to regain the level of permissible values.

1. INTRODUCTION

Several sources of heavy metals in the environment are anthropogenic in nature. Besides, heavy metal content in non-polluted soil heavily depends on the rocks from

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which the soil parent materials were driven. Meanwhile, the weathering process is quite relevant [1].

The presence of metals in trace amounts in the environment is of a fundamental importance. Some metals such as iron, manganese and zinc are considered to be nutrient elements for plants and thus play an important role in biochemical cycles [2]. On the other hand, some metals such as mercury, cadmium and lead are toxic to plants [3].

The use of pesticides, both organic and inorganic, in agriculture has been extended greatly after the World War II. This was in response to the concerted efforts for high yield of agricultural crops [4]. Pesticides form one of the input man-made sources of heavy metals in the agro-ecosystem in Egypt [5]. Thereby, the setbacks of using inorganic pesticides are due to the chemical and physical characters as well as toxicity of these compounds. Their strong resistance is a common property, which might be considered an advantage. Numerous cases of poisoning in men and animals were recorded after consuming pesticide-treated crops [4]. Thus, pesticide-born heavy metals are extremely hazardous when they find their way to surface and/or ground water [4], [5].

The major part of heavy metals is usually absorbed in silt [3]. It was revealed that heavy metals reaching the soil undergo several interactions with different soil constituents as well as with organic matter which affects both their mobility in the environment and their availability to plants [6]. It was reported that high levels of heavy metals in soil are detrimental to plants [7], [8]. Among the adverse effects is the retardation of growth resulting in stunted plants.

Many authors reported that the greatest amount of heavy metals taken up by plant was accumulated mostly in the root, and the least amount was found in the edible portion [9]–[11].

The wide misuse of fungicide-containing heavy metals in some developing countries deserves careful and comprehensive investigation. Mancopper is one of the commonly used fungicides in Egypt. The current study aimed to investigate the paths as well as the impact of this fungicide on the environment. Accumulation and translocation of metals in both plant and soil over a period of successive seasons were also evaluated. Furthermore, pot experiments were carried out for the sake of comparison with a particular regard to the level of metals in the drain water. The study was carried out at variable and controlled doses.

2. . MATERIALS AND METHODS

The use of Mancopper fungicide for the protection of potato crop in an Egyptian village named Qatta is the subject of the present study. The village is located 20 km southwest of Cairo in Giza Governorate. Its total population approaches 450–500. The main activity of the people is farming, while potatoes are among the main agricultural crops. The present study was carried out for a period of four successive seasons using

Mancopper fungicide. After cultivating the potato crop, water, soil and plant samples were monthly collected to investigate the level of such metals as zinc, iron, copper, manganese and lead. Similar samples were collected from a controlled field in the Nile Delta without using any fungicide for comparative purposes.

2.1. POT EXPERIMENT

Pot controlled experiments were carried out to investigate the response of plants to spraying them with fungicide in terms of the level of metals in plant, soil and drain water. In this experiment, the fungicide was applied in two different doses, i.e. field dose (2.5 g/dm^3) and double field dose (5 g/dm^3). The volume of each spray used was exactly 30 cm^3. Therefore, the total concentration of the fungicide applied reached 75 mg and 150 mg, respectively.

The fungicide was applied regularly every 15th day. Each pot of 50 cm height was filled with 20 kg of soil. The pot experiments were carried out in replicates. A control pot experiment was carried out in a similar manner without the fungicide.

2.2. IRRIGATION WATER SAMPLES

The samples of water used for irrigation in Qatta village, namely, the Nile and ground water were monthly collected for heavy metals determination according to U.S. EPA procedure and APHA [12], [13]. Water samples were filtered through Whatmann No. 4, then acidified by using concentrated nitric acid to pH below 2. The Nile water was used for irrigation purposes in the pot experiments.

2.3. SOIL SAMPLES

Field experiments were carried out on 6 plots, each of 5 m × 5 m. The surface soil samples (0–30 cm depth) were collected from three different locations, each irrigated with the Nile and ground water, as well as from the pot experiments. All locations were planted with potatoes and treated with Mancopper fungicide. Soil samples were oven-dried at 105 °C for 24 h, ground and passed through 100 μm sieve. Afterwards, weighed soil samples were acid-digested according to APHA [13] for metal analysis.

2.4. PLANT SAMPLES

Plant samples were collected from each location in duplicates and separated into root, stem, leaf and tuber. The samples were then oven-dried at 105 °C for 24 h and ground in blender. Then weighed plant samples were acid-digested according to APHA [13].

Soil texture, chemical and physical characteristics

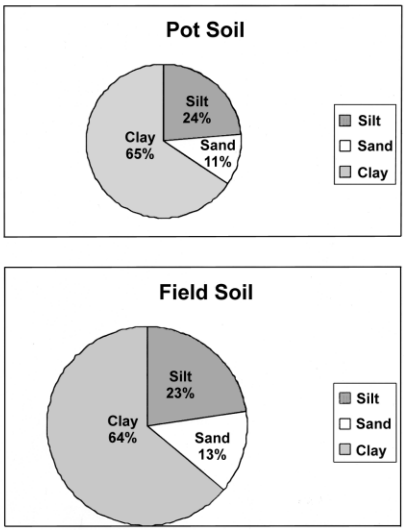
Location	Sand %	Clay %	Silt %	O.M. %	pH	N		P	
						Meq/dm^3		Meq/dm^3	
						Total	Sol.	Total	Sol.
Qatta	12.9	64	23.1	7.1	7.8	13.66	8.88	86.86	2.86
Pot	11.0	65.5	23.5	4.71	6.71	15.05	20.46	107.8	3.21

2.5. METAL INVESTIGATION

Metal concentrations were determined using Perkin-Elmer Atomic Absorption Spectrophotometer, Model 2380. The Mancopper fungicide was analyzed for the metal content. The tabulated result for each sample is an average of 10 sequential readings. Blanks for redistilled water were also acid-digested according to the procedure previously described. All present results are the mean values of the replicates.

3. RESULTS AND DISCUSSION

Mancopper fungicide was found to be composed of Zn, Fe, Cd, Cu, Mn and Pb at the respective levels of 179.75, 4.8, 2.55, 683.89, 39.5 and 0.9 mg/g as dry weight. Some physical and chemical characteristics of the soil that were used for the present investigation as well as the soil that was used for the pot control experiments are given in table 1 and the figure.



Soil texture in both field and pot experiments

Table 1

of soil from Qatta village and pot experiment

K		EC mS/cm at 25 °C	Ca^{2+} Meq/dm^3	Na Meq/dm^3	Mg^{2+} Meq/dm^3	CO_3^{2-} Meq/dm^3	HCO_3^- Meq/dm^3	Cl^- Meq/dm^3	SO_4^{2-} Meq/dm^3
Meq/dm^3									
Total	Sol.								
8.86	8.83	0.22	1.05	0.18	0.86	1.07	1.52	0.13	0.28
10.5	9.17	0.39	1.56	0.20	1.21	1.15	1.78	0.28	0.38

O.M. – organic matter.
Sol. – soluble.
E.C. – electric conductivity.

3.1. LEVEL OF METALS IN IRRIGATION WATER

The level of metals in the water used for irrigation in three successive seasons is given in table 2. The results obtained show that the level of metals is within the permissible range according to the Egyptian regulation [14] as well as the WHO guidelines [15]. It is worth noting that in the summer the levels of the metals recorded were slightly higher than those in other seasons, particularly for zinc and copper. This may be attributed to the seasonal flood of the Nile in August, which enriched the river water with clays, minerals and organic matter as a result of the running off process [16].

Table 2

Level of metals in two types of water used for irrigation as well as in the drain water in the field of Qatta village during a period of three seasons studied (mg/ dm^3). The plants were not sprayed with fungicide

Type of water	*n*		Zn	Fe	Cd	Cu	Mn	Pb
The Nile water		Min	0.019	0.112	0.000	0.021	0.034	0.005
		Max	0.067	0.357	0.004	0.058	0.83	0.033
	12	Mean	0.048	0.278	0.003	0.048	0.069	0.023
Ground water		Min	0.011	0.055	0.000	0.09	0.028	0.008
		Max	0.042	0.304	0.000	0.043	0.078	0.037
	12	Mean	0.025	0.116	0.000	0.021	0.059	0.019
Drain water		Min	0.092	0.271	0.002	0.033	0.035	0.021
		Max	0.210	0.750	0.012	0.126	0.159	0.070
	12	Mean	0.120	0.502	0.008	0.097	0.095	0.048
Egyptian permissible limits			2.0	5.0	0.01	0.20	0.20	5.0

n – number of samples.
Mean – a mean value.

Meanwhile, the minimum values were recorded mostly in the cold season, i.e. winter, which is in agreement with previous studies [5], [17]. On the other hand, the average levels of metals in the ground water are almost similar to those in the Nile water. This finding may throw light on the proportionate enter-correlation between both sources. However, no significant changes were observed seasonally in the ground water composition which testifies to the stability of this type of water.

3.2. FIELD LEVEL OF METALS IN SOIL

The control samples of soil were collected from pots where plants were grown without any fungicide addition. A mean level of metals in the soil from Qatta village showed no serious contamination. Nevertheless, iron, copper, lead and cadmium reached their highest levels in the summer season, whilst zinc and manganese approached their maximum levels in autumn and summer, respectively. It is worth noting that no additional contribution of heavy metals to the soil was due to soil irrigation with either the Nile water or ground water (table 3). Alternatively, it can be inferred that a prolonged use of ground water for irrigation increases the soil salinity [18].

Table 3

Level of metals in the field soil of Qatta village at the depth of 0–30 cm and in the pot soil (mg/kg of dry weight)

Type of soil	*n*	Zn	Fe	Cd	Cu	Mn	Pb
1. Qatta field soil Irrigated with the Nile water*	12	58	356	1.20	32	54.2	43
Irrigated with the Nile water**	12	60	357	1.23	41	55.3	4.3
Irrigated with groundwater*	12	47	324.8	1.21	47	82.2	38
Irrigated with groundwater**	12	50	326.1	1.23	55	83.0	3.9
2. Pot soil Irrigated with the Nile water*	12	30	184	0.66	17	36.1	2.4
Irrigated with the Nile water**	12	33	185	0.69	26.5	37.8	2.4
Irrigated with the Nile water***	12	37	189	0.72	32.7	39.3	2.5

n – number of samples.
* Without any fungicide addition.
** After the application of fungicide (field dose of 2.5 g/ dm^3).
*** After the application of fungicide (double field dose of 5.0 g/ dm^3).

In the field experiment, the plants were sprayed with the fungicide in the dose of 2.5 g/dm^3 and watered with the Nile and ground water. In these circumstances, a slight increase in the metal concentration in soil was detected; this concentration was compared with that in the control soil (table 3). An increase in the level of copper, zinc and manganese can be ascribed to their relatively high content in the Mancopper. The present finding is in agreement with these reported by ABDEL-SHAFY et al. [4], [18] and BADAWY [19].

3.3. HEAVY METALS IN POTATO PLANT

Table 4 presents the level of heavy metals in different parts of potato plants irrigated with Nile and ground water for over four successive seasons in Qatta village. These levels can be compared with the levels in untreated parts of the plants grown in the controlled pot experiments (table 5). As expected, the comparison proves that the samples treated with fungicide (table 4) demonstrate slightly higher concentration of the metals tested than the untreated ones (table 5). In the period of continuous soil irrigation with both the Nile and ground water, most of metals, namely iron, cadmium, manganese and lead, are accumulated in plants. If their concentrations in potatoes are taken into account, the parts of the plant can be arranged in the following ascending order: leaf > root > stem > tuber. Comparing the level of metals in the plant grown in the field with that of plants grown under controlled conditions, it can be seen clearly that use of the fungicide increased the levels of copper, zinc and manganese followed by iron. Virtually, in the edible plant part, i.e. in the tuber, accumulation of metals was the lowest compared to the other plant parts. On the other hand, iron showed a remarkably higher concentration than the other heavy metals in the plant.

Table 4

Mean contents of heavy metals (mg/kg of dry weight) in different parts of potato plants irrigated with the Nile and ground water in the field of Qatta village at a 2.5 g/dm^3 dose of fungicide

Metal	Water sources	*n*	Root	Stem	Leaf	Tuber
Zn	N.W.	12	46	38	51.66	19.33
	G.W.	12	31.66	21.33	49	14.66
Fe	N.W.	12	111.66	79.66	97.33	62.66
	G.W.	12	109.66	62.66	93.33	61
Cd	N.W.	12	0.8	0.5	1.7	0.26
	G.W.	12	0.6	0.4	1.66	0.23
Cu	N.W.	12	16.4	13.9	53.33	2.56
	G.W.	12	16.3	13.9	53.33	2.3
Mn	N.W.	12	46.9	36.66	54	28
	G.W.	12	57.33	36	53	29.33
Pb	N.W.	12	3.8	0.66	4.13	1.93
	G.W.	12	3.9	0.66	3.6	1.90

N.W. – the Nile water.
G.W. – ground water.
n – number of samples.

Table 5

Mean contents of heavy metals (mg/kg of dry weight)
in different parts of potato plants grown in pots and irrigated with the Nile water

Metal	*n*	Root	Stem	Leaf	Tuber
Zn	9	29	20	36	13
Fe	9	102	61	81	44
Cd	9	0.9	0.4	0.5	0.2
Cu	9	3.0	2.3	2.6	1.4
Mn	9	37	30	32	23
Pb	9	2.3	0.6	1.7	1.1

n – number of samples.

Zinc level in the tubers of potatoes irrigated with both the Nile water and/or ground water was definitely lower than the minimal threshold commonly ranging from 25 to 125 mg/kg [20]. It has been reported by SILLANPEA [21] that the level of copper in the plant leaves < 4 mg/kg testifies to its deficiency, while the concentration from 6 to 20 mg/kg is sufficient, and the concentration higher than 20 mg/kg is toxic to plants. Thus, the samples examined revealed substantially toxic copper levels in the plant leaves (table 4). In the case of an intermediate iron content, its mean level in the samples studied is considered to be normal. Mean levels of cadmium are similarly normal. BINGHAN et al. [22] have found that the cadmium concentration of 20 mg/kg is critical for plant tissues, and a higher one is toxic. Therefore, the level of cadmium in the present study is considered to be within the permissible values. Manganese concentration in the dry matter of plants grown in soil usually ranges between 18 and 55 mg/kg [23]. Hence, manganese mean levels in the roots and leaves of collected samples can be considered slightly higher or within a normal range of concentration (table 4). Lead concentration may testify to its substantial accumulation in the samples collected. Previous investigations have shown that a typical lead level in grain and other plants in non-polluted areas are usually less than 2.0 mg/kg [24], [25]. The level of lead in our investigation (tables 4 and 5) was even less than that reported by the same authors [24], [25]. This can be attributed to its low level in Mancopper.

3.4. POT EXPERIMENT

The plants in pot experiment were treated with two different doses of Mancopper, i.e. 2.5 g/dm^3 and 5.0 g/dm^3, which were equivalent to the total concentration of fungicide reaching 75 and 150 mg, respectively (table 6). A remarkable increase in the level of metals in the leaves was similar to that in the control plants (tables 5 and 6). At a 2.5 g/dm^3 dose of the fungicide the increasing factor of metals in the plant leaves reached the value of 1.61, 1.88 and 3.77 for zinc, lead and copper, respectively, while at a 5.0 g/dm^3 dose it assumed the value of 2.11 for zinc, 2.59 for lead and 6.75 for copper. These results may again reflect the concentration of the metals in the fungicide applied (tables 5 and 6).

Table 6

Mean contents of heavy metals (in soil and in different parts of the plant) after the application of two different doses of the Mancopper (2.5 g/dm^3 and 5.0 g/dm^3)

Sample	Field concentration dose (2.5 g/dm^3)* Metal (mg/kg of dry weight)						Double field concentration dose (5.0 g/dm^3)** Metal (mg/kg of dry weight)					
	Zn	Fe	Cd	Cu	Mn	Pb	Zn	Fe	Cd	Cu	Mn	Pb
D.W. (mg/dm^3)	0.24	0.9	0.009	0.23	0.16	0.055	0.41	1.72	0.015	0.437	0.21	0.068
Soil	33	185	0.69	26.5	37.8	2.4	37.0	189.1	0.72	32.7	39.3	2.5
Root	34	165.4	1.2	5.0	39.25	2.54	36.4	177	2.40	9.1	47.49	2.9
Stem	29	66.4	0.62	5.6	41	0.8	36.7	75	1.1	11.08	47.43	1.8
Leaf	58	153	1.8	9.8	54	3.2	76	185	2.63	17.55	62	4.4
Tuber	16	61	0.31	2.60	28	1.2	27.3	77	0.5	5.1	36	1.34
Level of elements tolerated by plant +	10–50	100	0.1–1.0	5.0–20	80	0.1–5	10–50	100	0.1–1.0	5.0–20	80	0.1–5

* Equivalent to the total of 75 mg (for each one spray).

** Equivalent to the total of 150 mg (for each one spray).

D.W. – Drain water.

+ Egyptian Association of Environmental Affair.

For tuber, the edible part of the plant, the results obtained revealed that the increasing factors were at their minimum compared with the factors calculated for the other parts. Their values were as follows: 1.09 for lead, 1.23 for zinc and 1.86 for copper at the Mancopper dose of 2.5 g/dm^3. The corresponding factors increased to 1.22, 2.10 and 3.64 for lead, zinc and copper at the fungicide dose of 5 g/dm^3 (tables 5 and 6). It is worth noting that the levels of metals in the tuber at both doses are within the permissible limits according to both FAO and the Egyptian limits [14], [26] and far less than these reported for contaminated areas [27].

In the case of roots and stems, the increasing factors were much less than the corresponding factors for leaves (tables 5 and 6). This may be attributed to the fact that the leaves only were sprayed with the fungicide, thus it has no effect on the roots.

Due to the field treatment (2.5 g/dm^3) the content of all metals tested in soil (table 6) increased slightly. The increasing factors were: 1.10, 1.56 and 1.05 for zinc, copper and manganese, respectively. Alternatively, for a double field treatment (5 g/dm^3) the increasing factors were: 1.23, 1.92 and 1.09 for the mentioned metals, respectively. It is worth mentioning that the highest concentration factors were recorded for copper, while the lowest were for lead and cadmium (tables 3 and 6). This can be explained by the fungicide composition.

There was observed a noticeable increase in the metal contents in drain water at both field and double-field doses of Mancopper (table 6) compared with irrigation water (table 2). Nevertheless, the metals in the drain water at both doses were still within the permissible levels recommended by WHO guideline and the Egyptian regulations [14], [15].

4. CONCLUSIONS

- The level of heavy metals in different water sources can be arranged in the following order: ground water < the Nile water, and both are still within the permissible levels.
- The level of heavy metals in soil does not depend essentially on the sources of irrigation, but first of all on the metal constituents of the fungicide applied.
- The leaves being sprayed with fungicide showed a remarkable increase of in copper, zinc and manganese concentrations.
- The accumulation of heavy metals by plant parts varied seasonally and depended mainly on the fungicide applied.
- Despite a relatively high dose of the fungicide applied the edible part, tuber, was still not jeopardized.
- Pesticides impose an adverse impact on the environment, water, soil and crops. They should be used for plant disease control in proper and least possible concentrations.
- Soil should carefully be washed to decrease the metal concentration. Bioremediation methods can be also recommended in this respect.

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MIGRACJA METALOIDOWYCH FUNGICYDÓW W WODZIE, GLEBIE I ROŚLINACH W EGIPCIE

Metale ciężkie należą do jednych z najbardziej uciążliwych zanieczyszczeń środowiska. Jednocześnie są szkodliwe dla zdrowia człowieka. Pierwiastki te występują w wielu przemysłowych produktach, np. w nawozach sztucznych i pestycydach. Łatwo przenikają do wód powierzchniowych i, w konsekwencji, do łańcucha pokarmowego.

Celem badań było określenie zawartości metali ciężkich w wodzie, glebie i uprawianych ziemniakach po zastosowaniu środka grzybobójczego Mancopper w testach polowych i laboratoryjnych (testy doniczkowe). Fungicydu o nazwie Mancopper używano w okręgu Giza w Egipcie. Eksperyment prowadzono przez cztery sezony uprawne, określając w tym czasie średni poziom takich pierwiastków jak: Fe, Zn, Mn, Cd i Pb. Stwierdzono, że poziom metali ciężkich w warstwie gleby nawadnianej wodą z Nilu malał następująco: Fe > Zn > Mn > Pb > Cu > Cd. Z kolei stężenie tych pierwiastków w glebie nawadnianej wodą podziemną malało zgodnie z szeregiem: Fe > Mn > Zn = Cu > Pb > Cd. Zawartość metali ciężkich w częściach roślin wyhodowanych na nawadnianej glebie obniżała się w następujący sposób: korzeń > liść > łodyga > bulwa. Stwierdzono, że dopuszczalne stężenia miedzi i ołowiu w jadalnych częściach ziemniaków (w bulwach) zostały przekroczone. Okazało się również, iż mimo małego stężenia ołowiu w środku grzybobójczym, zawartość tego pierwiastka w glebie była znaczna. Obserwacje te świadczą o migracji metali w glebie i ich akumulacji w roślinach.