

Pesticide Contamination of Surface Water in Egypt and Potential Impact

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ABSTRACT

Enzyme-immunoassay field kits and capillary gas chromatography with nitrogen-phosphorous and electron capture detection (NPD and ECD) systems were used to determine the levels and distribution of commonly used pesticides (alachlor, atrazine, butylate, cyanazine, diazinon, metolachlor, metribuzin, malathion, parathion, propachlor, pendimethalin, trifluralin) and some of their metabolites (diethyl aniline, hydroxy alachlor, DE-atrazine, and DIP-atrazine) in surface water in Egypt. Replicate water samples were collected from several sampling sites near Cairo, Alexandria, Damietta, and Manzala Lake, using solid-phase concentration onto C₁₈ cartridges, and the extract was subsequently analyzed using GC/NPD and GC/ECD systems. Alachlor, metolachlor, trifluralin, parathion, malathion, atrazine, and metribuzin were the most frequently detected pesticides, in the order listed, at the four sampling locations. Alachlor had the highest concentration detected (165-254 ppb), followed by metribuzin (0.6-47 ppb), metolachlor (6.6-12.5 ppb), atrazine, (2.5-7.3 ppb), and malathion (1.5-5.7 ppb). The concentrations of pesticides detected at Cairo were generally lower than those at the other three locations, down stream on the Nile River. The largest number of pesticides detected was in the Damietta branch of the Nile, and their concentration was generally higher than those detected at all other locations. Manzala Lake is the second highest location in terms of overall frequency and concentration of pesticides detected. The ecological and environmental health impacts of such contamination will be discussed.

Key words: Pesticides, water, Manzala, Damietta, Cairo, Alexandria.

INTRODUCTION

The intensive three-crop agricultural cycle in the Nile Delta has become heavily dependent on the application of large quantities of pesticides. Over 300 trade name pesticides, manufactured by over 50 chemical companies, are currently used in Egypt (Selim and El-Sebae, 1995). During the period from 1984 - 1988, a total of 41,368 tons of insecticides, 17,854 tons of fungicides, and 12,021 tons of herbicides were imported into Egypt. The imported pesticides include the main organophosphorous, carbamates, and pyrethroids groups. The use pattern showed a positive increase in the use of insecticides (56.4% to 70.5% of total-imported insecticides) and herbicides (21.3 to 28%) between 1984 and 1988 (Maklad, 1989). With the continuous population growth and the need for increasing agricultural production, more pesticides are expected to be imported and used in Egypt. The majority of these pesticides are used to control pests and weeds in cotton, wheat, maize, rice, sugar cane, vegetables, and soy bean crops.

Aerial spray of pesticides (which has stopped years ago) resulted in residues finding their way to humans, animals, and fish. However, manual application has also resulted in many documented cases of acute poisoning to humans (El-Wafd, 1993), farm animals, honey bees (El-Sebae, 1990), and fish (El-Akhbar, 1991), due to direct dermal contact or ingestion of contaminated food, water, or feed occurring annually in Egypt.

Organochlorine pesticides such as endrin, dieldrin, heptachlor, DDT, and lindane have been detected in marine organisms (Ernst *et al.*, 1983), food samples (Abdel-Gawad and Shams El-Din, 1989, 1990; Dogheim *et al.*, 1988, 1991), surface water (Fathi *et al.*, 1990; Khalifa, 1991), and agricultural drainage water and soil (Maklad *et al.*, 1990; El-Gendy *et al.*, 1991). A previous study showed that drinking water treatment had partial effect on the levels of chlorinated pesticides in surface water (El-Sebae *et al.*, 1989). Low levels of DDE and DDT were totally removed by the water treatment, but HCH are not due to their greater solubility in water.

Detection and persistence of the organochlorine pesticides in the Egyptian environment has discouraged their use and supported their gradual substitution with less stable, and presumed "safer pesticides." such as organophosphorous, carbamates, and synthetic pyrethroids. Organophosphorous pesticides were introduced in early 1970's followed by synthetic pyrethroids in 1977 (Khalifa, 1990). In 1985 for example, 1,000 tons of organophosphorous insecticides, 800 tons synthetic pyrethroids, and 550 tons carbamates were imported into Egypt (El-Sebae, 1990). These quantities are expected to increase over the last ten years, due to their increased availability and acceptability as a substitute to organochlorine pesticides. The perceptions of "safer" pesticides have lead to their extensive application and misuse which resulted in the development of 'pest resistance' (Khalifa, 1990; Selim and El-Sebae, 1995). To overcome "pest

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resistance” farmers resorted to multiple pesticide applications or formulating their own mixtures of pesticides. Many common name pesticides such as, alachlor and atrazine, became available as mixtures under different trade names (Selim and El-Sebae, 1995), which make it difficult to estimate the level or pattern of usage for a given compound. However, there has been a total lack of data on their levels or distribution in the Egyptian environment. Available literature has focused only on the neurotoxicity of organophosphorous pesticides (El-Sebae *et al.*, 1977, 1980, 1988) and their residue in some crops such as onions (Hegazy *et al.*, 1988) sugar beet roots and tops (Kady and Przezdziecki, 1985). Studies of pesticide residue in surface water in Egypt have focused mainly on organochlorine pesticides (Hassan *et al.*, 1996; Osfor *et al.*, 1998; Abbassy, 1999; Abbassy *et al.*, 2000; Gupta *et al.*, 2003). However, to the best of our knowledge, monitoring data for pesticides other than the organochlorine (endrin, dieldrin, lindane, and DDT and its metabolites DDE and DDD) group has been extremely rare or non existent.

Therefore, the primary objective of this preliminary study was to investigate the problem of pesticide contamination of surface water in Egypt, particularly in relation to pesticides that are widely used and for which monitoring data is lacking, such as alachlor and triazines.

MATERIALS AND METHODS

Sampling

Three sampling locations were selected along the River Nile, one at Cairo and at its lower most parts and output canals at Alexandria and Damietta (Fig. 1). A fourth sampling location was selected at Manzala Lake, which is the receiving body for agricultural drain water. Multiple sampling sites were also selected at each sampling location; three at Cairo (population =17 million), four at Alexandria (population = 4.4 million), eight at Damietta (population = 1.1 million), and two at Manzala Lake (Egyptian Environmental Affairs Agency, 2006). At Cairo, one sampling site was for treated water at El-Mohandsein and two sampling sites were for raw river water. One at the University of Cairo bridge, the second was approximately one Kilometer north of Embaba bridge, and the fourth was for treated water at Heliopolis. At the Alexandria location, the treated water sampling site was at Alexandria University, Department of Pesticide Chemistry. Two raw water sampling sites were located at El-Mahmoudeya Canal, which is the main intake for the water treatment plant for Alexandria City. Three treated water sampling sites at the Damietta location were located at Kafr El-Battikh City, at Raas El-Barr resort, and at downtown Damietta City. The four raw water sampling

sites were located at the intake of the water treatment plant at Kafr El-Battikh, at the intake canal for the Damietta water treatment plant (at Welad Hamam), and on the river above and below the isolation dam at Damietta (seals off the fresh water, approximately 15 miles before the Mediterranean). The Manzala Lake location had two sampling sites of raw surface water, one at El-Litha and El-Gameal. The four raw water sampling were taken as close as possible to the center of the sampled river or the canal and approximately one meter below the surface. A minimum of two replicate samples for pesticide analysis was collected at each sampling site.

Materials and Reagents

Pesticide standards were 99+% pure, obtained from ChemServices (West Chester, PA) or the USEPA (Research Triangle Park, NC). All solvents were J.T. Baker pesticide residue grade (J.T. Baker Chemical Company, Phillisburg, N.J.). Pesticide standard solutions were made up in isooctane. All pesticide solutions were stored in amber glass vials, sealed with Teflon tape, and stored in the dark at -4°C. The solid phase extraction cartridges were Waters Sep-Pack tC18 (Environmental), containing approximately one gram of the packaging material (Waters, Division of Millipore, Bedford, MA). All glassware was cleaned with water and laboratory detergent, rinsed with methanol followed by acetone, and dried at 100°C. The use of plastic containers or septa was avoided, except for Teflon coated parts.

Enzyme Immunoassay

The immunoassay kits used for the field and laboratory screening were EnviroGard Triazines and Cyclodienes tube-Kits donated by Millipore Corporation (Bedford, MA). The Triazines Kit tests for atrazine, Simazine, propazine, prometryn, ametryn, simetryn, cyanazine, terbutylazine, trietazine, prometon, 6-hydroxy atrazine, DE atrazine, DIP atrazine, and diazinon. The Cyclodienes kit tests for endrin, chlordane, dieldrin, aldrin, heptachlor, and endosulfan. The kits are designed to be used for semiquantitative screening in conjunction with a portable differential photometer. The kits were used following the manufacturer procedures, which are provided in the kits inserts. More details of the kits procedure and their performance relative to GC/MS analysis are published specific to kits and are published elsewhere (Selim *et al.*, 1995).

Solid Phase Extraction

The cartridges were first conditioned by consecutively passing 5, 5, and 10mL of ethyl acetate, methanol, and water, respectively. A 1 L water sample was then passed through the cartridge at a flow rate of 15-20mL/ minutes, using a 30 ml glass

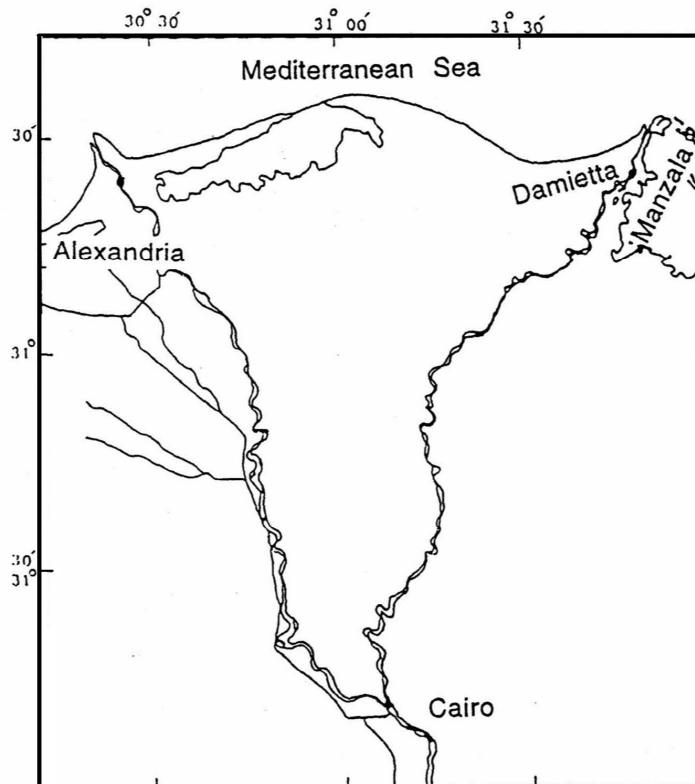


Figure (1): Sampling Locations.

syringe. After the water sample was passed, the cartridge was dried by passing dry air through it for approximately 10-15 minutes, using a dry glass syringe. The cartridges were sealed from both ends, placed in ziplock bags and kept cold on ice during transportation, and refrigerated until ready for analysis. Upon receipt of the samples at the University of Iowa, the extract was eluted by slowly passing approximately 3.5mL ethyl acetate (Nash, 1990; Johnson *et al.*, 1991; Cai *et al.*, 1993). The extract was passed over approximately 0.5 gram of anhydrous sodium sulfate and collected into a 13-mL Graduated conical tube. The final extract was evaporated by nitrogen blowing and its final volume was adjusted to 1mL.

GC Analysis

Qualitative and quantitative chromatographic analyses of organophosphorous pesticides were carried out using a 5890 GC, Series II (Hewlett -Packard, Palo Alto, CA) equipped with two nitrogen phosphorous detectors and two capillary columns; a 0.53 mm x 30 m DB-17, 1 mm film, and a 0.53 mm x 10 m, 1.33 mm Carbowax 20M (Alltech Associates, Deerfield, IL). HP3365 ChemStation software and MS Windows on a 386 Gateway2000 computer (Gateway2000, Sioux City, ND) were used for data collection and analysis. Chlorinated pesticides were analyzed using a Varian model 3400 GC (Varian, Sugar Land, TX) equipped

with two electron capture detectors and two capillary columns; a 0.25 mm x 30 m, 0.25 mm film DB-210, and a 0.25 mm x 30 m, 0.25 mm film DB-17. This GC was connected to Spectra-Physics SP4270/SP4290 integrators and LABNET computer system (Spectra-Physics, San Jose, CA) for data collection and analysis. Qualitative identification of pesticides in the test samples was based on comparing their relative retention with that of known standard. Quantitative determination of the concentration was based on the method of internal standard with multilevel calibration. Triphenylphosphate (TPP) was used as the internal standard for compounds analyzed by NPD and pentachloronitrobenzene (PCNB) for compounds analyzed by the ECD. Field blanks and spikes were used to account for sample stability in transit and matrix effects. These methods were based on The United States Environmental Protection Agency Methods for the Analysis of Organic Compounds in Drinking Water and their adaptations, which are published elsewhere (USEPA, 1991; Kross *et al.*, 1992; Selim *et al.*, 1996).

RESULTS AND DISCUSSION

A list of pesticides targeted for this investigation is provided in Table (2). These pesticides were selected based on their widespread use in both Egypt (Selim and EI-Sebae 1995), as well as in the US, (Kross *et al.*, 1992) and the availability of established GC methods

for their determination and analysis (USEPA, 1991; Kross *et al.*, 1992; Selim *et al.*, 1996). Enzyme immunoassay (EnviroGard Kits) was used to screen for the presence of the cyclodiene pesticide group and for triazine pesticide group, which include the most commonly used nitrogen containing pesticides. Because of difficulties in voltage transformation (220/110 V) for the field equipment, initial field screening was qualitative and relied on visual detection of the color change. Both pesticide groups (triazines and cyclodienes) visually showed a high positive reaction at all field sampling locations. These results were later confirmed in the laboratory using a differential spectrophotometer. Since chlorinated pesticides detected by the cyclodiene immunoassay kit (endrin, chlordane, dieldrin, dieldrin, aldrin, heptachlor, and endosulfan) and have been previously measured at the same sampling sites using GC/ECD methods, (El-Sebae, 1989; Maklad, 1990; El-Gendy, 1991) no further

confirmation was conducted for these pesticides. Figure (2) shows the chromatographic separation of the pesticides listed in Table (2) on the DB Carbowax 20M with the NPD system. Similar separation was obtained with the DB-210 column with the ECD system, except for the enhanced detection of the electron capturing compounds. A summary of the GC quantitative analysis for the samples analyzed at the four sampling locations is presented in Table (3). The types of pesticides detected and the frequency of their detection at each sampling location are also presented in Figure (3). As shown in Figure (3), alachlor, metolachlor, trifluralin, parathion, malathion, atrazine, and metribuzin were detected at all four sampling locations. The frequency of their detection follow approximately the order listed, with alachlor having the highest frequency of detection at all sites. The mean concentrations of detected pesticides are compared in Figure (4). In this figure, log mean

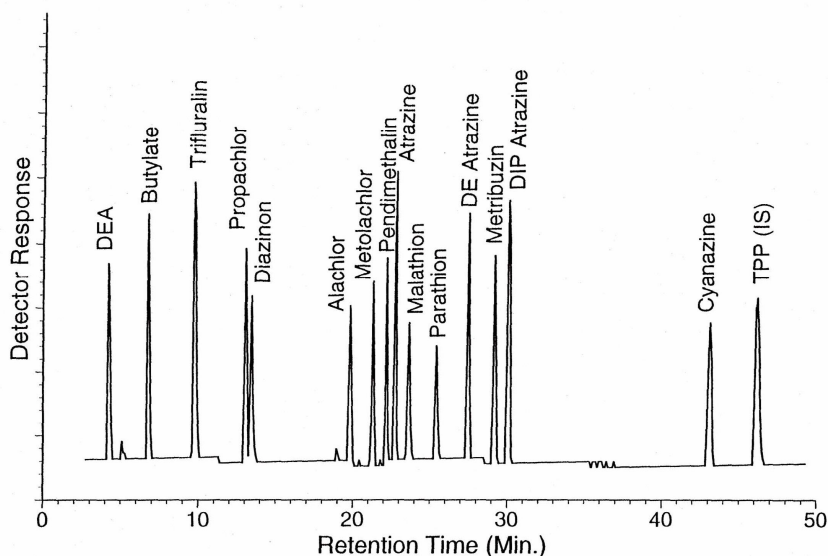


Figure (2): Separation of Pesticides on the DB-Carbowax 20M column, NPD.

Table (1): GC/NPD Parameters (Varian 6000), and GC/ECD Parameters (Varian 3400).

GC/NPD Parameters (Varian 6000)		GC/ECD Parameters (Varian 3400)	
Channel A	Channel B	Channel A	Channel B
DB-17*	Carbowax 20M†	DB-210†	DB-17*
1 µm film	1.33 µm film	0.25 µm film	0.25 µm film
30m x0.53 mm ID	10m x0.53 mm ID	30 m x 0.25 mm ID	30 m x 0.25 mm ID
Conditions		Conditions	
Injection Temperature: 225 °C		Injection Temperature: 220 °C	
Detector Temperature: 300 °C		Detector Temperature: 275 °C	
Oven Temperature: 130 °C for 5 min., 4 °C/min., 184 °C		Oven Temperature: 140 °C for 5 min., 4 °C/min., 200 °C for 7 min.	

*Product of Alltech Associates, Deerfield, IL

Table (2): Pesticides selected for this study.

Commercial Name	Common Name (Metabolite)	Use
Aatrex, Atrazine Basudin	Atrazine (DE-, DIP, & OH-Atrazine) Diazinon	Selective herbicide –corn and sorghum for weed control Insecticide – citrus, apple, peas, & sugar beet
Bladex, atrazine	Cyanazine, atrazine	Selective herbicide
Lasso	Alachlor (DEA, OH-Alachlor	reemergence herbicide -soybeans, corn, peanuts
Lasso/Atrazine	Alachlor/Atrazine	Insecticide - maize
Linuron/Alachlor	Linuron/Alachlor	Insecticide - soybean
Lexone, Sencor	Metribuzin (DA, DADK,DK)	herbicide - agricultural crops
Malathion	Malathion	Insecticide- Cabbage, calif lower, artichoke, potato, beans, squash, cucumber, melons, citrus, date palm, guava, mang, figs, maize,wheat,
Prowl	Pendimethalin	herbicide - corn, potatoes, rice, sorghum, cotton, soybeans, and tobacco
Ramrod	Propachlor	Reemergence to early post herbicide
Stomp	Pendimethalin	Herbicide - cotton, onion, soybean, tomato, and rice
Sutan	Butylate	Selective preplant herbicide - corn
Treflan	Trifluralin	Reemergence herbicide - alfalfa, soybeans, cotton and vegetable crops

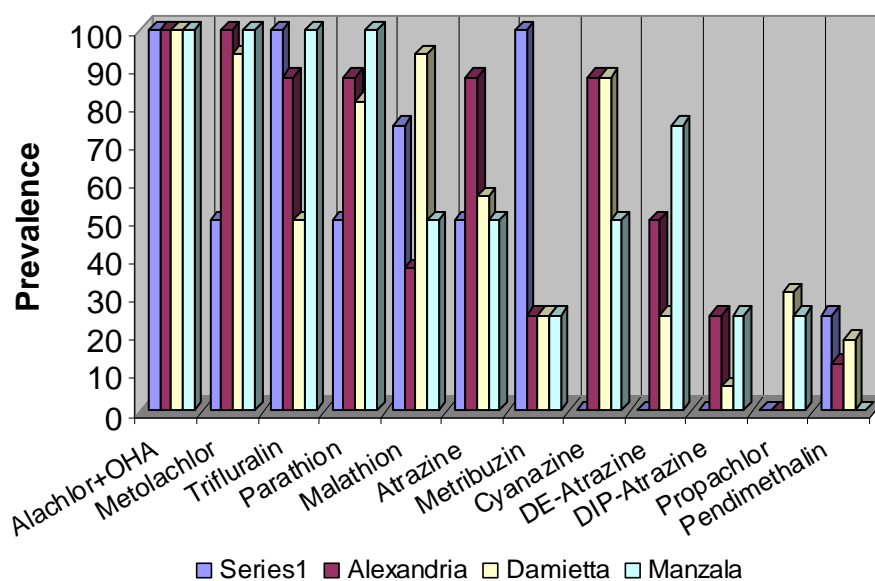


Figure (3): Plot comparing the prevalence of pesticides detected at the four sampling locations.

concentration is plotted for all pesticides detected at the four sampling locations. As listed in Table (3) and shown in Figure (4), alachlor had the highest mean concentration detected, followed by metribuzin and metolachlor. Among all sampling locations, samples collected from Manzala Lake showed the highest mean concentration of alachlor (254 µg/L). Trifluralin had the lowest mean concentration (0.06 -0.11 µg/L) at all locations followed by propachlor. Within the four sampling locations there was no significant difference between the concentrations of pesticides detected in raw water and their concentration in the treated drinking water at their corresponding sampling locations. This is consistent with previous study on the effect of water treatment on the removal of chlorinated pesticides from drinking water in Alexandria, Egypt (El-Sebae *et al.*,

1989; El-Gendy *et al.*, 1991). This indicates that the conventional water treatment steps (flocculation, sedimentation, and chlorination) used in Egypt are not capable of and were not designed to remove pesticide contaminants from the treated drinking water. Except for metribuzin, all pesticide concentrations measured at the Cairo location were lower than those measured at the three other locations, down stream of the intensive agricultural activities in the Delta. Metribuzin concentrations and frequency of detection were highest at Cairo and lowest at Manzala Lake. This possibly due to the greater use of metribuzin in upper Egypt or in the other countries up stream of Egypt. Since the river Nile originates from the African plateau and with its tributaries (6,825 km) cross eight countries (Sudan, Ethiopia, Uganda, Tanzania, Kenya,

Pesticide contamination in Egypt

Table (3): Summary of prevalence and concentrations of pesticides detected at the four sampling locations in Egypt.

Pesticide	Cairo		Alexandria		Damietta		Manzala	
	Prevalence (n=4)	Concn (ppb)	Prevalence (n=8)	Concn (ppb)	Prevalence (n=16)	Concn (ppb)	Prevalence (n=4)	Concn (ppb)
Alachlor+OHA	100.00	165.00	100.00	186.88	100.00	192.62	100.00	254.00
Metribuzin	100.00	47.02	25.00	24.16	25.00	33.79	25.00	0.64
Metolachlor	50.00	6.60	100.00	10.70	93.75	12.46	100.00	11.94
Atrazine	50.00	2.50	87.50	3.50	56.25	2.91	50.00	7.29
Malathion	75.00	1.76	37.50	7.63	93.75	5.73	50.00	1.51
Pendimethalin	25.00	1.80	12.50	0.68	18.75	4.11	0.0	0.0
Parathion	50.00	0.00	87.50	1.17	81.25	1.90	100.00	1.70
DE-Atrazine	0.00	0.00	50.00	0.63	25.00	1.50	75.00	1.46
DIP-Atrazine	0.00	0.00	25.00	0.80	6.25	0.68	25.00	0.76
Cyanazine	0.00	0.00	87.50	1.03	87.50	1.18	50.00	1.45
Trifluralin	100.00	0.06	87.50	0.11	50.00	0.06	100.00	0.11
Propachlor	-	-	-	-	31.25	0.57	25.00	0.42

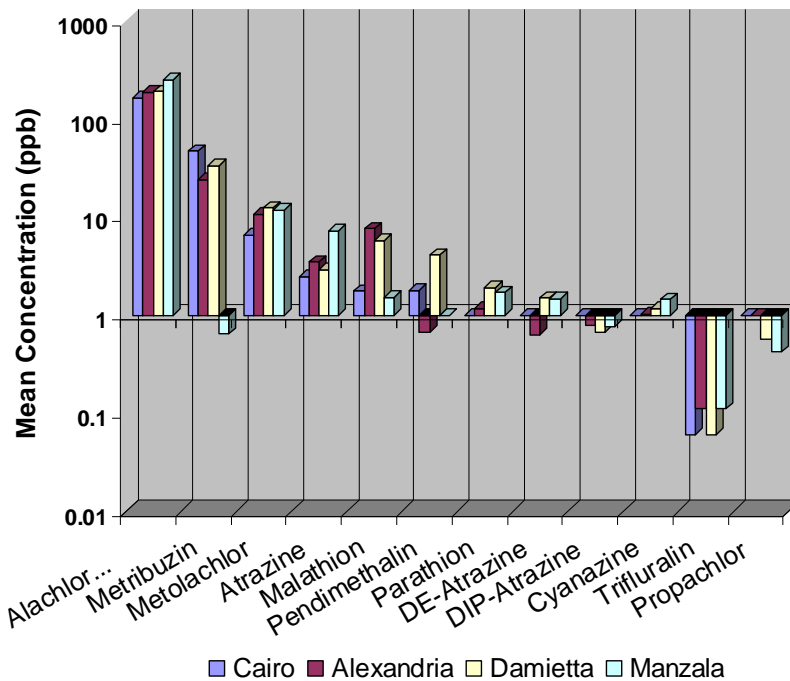


Figure (4): Mean concentrations of Detected Pesticides at the four Sampling Locations.

Zaire, Rwanda, and Burundi), it is difficult to assess if the source of contamination is from the agricultural activities in upper Egypt or from other countries upstream of the Egyptian territories. The Damietta branch of the Nile appears to be the most contaminated of the four sampling locations. All tested pesticides were detected in the Damietta branch. The frequency of pesticide detection and their concentration were generally higher than those at the other three locations. A similar trend was observed in a previous study for the concentration of chlorinated pesticides in the Damietta and Rosetta branches of the Nile (Fathi *et al.*, 1990) This is probably due to the high volume of agricultural drainage discharged to Damietta branch through two major agricultural drainage canals (El-Sooror El-Aala

and dishrag Canal #1). These two canals discharge a total of approximately 1,100,000 m³/day to the Damietta branch of the Nile at the City of Meet Ghamr and Kafr El-Dabboos (Damietta Department of Environmental Affairs, 1991). Manzala Lake, which receives the second major portion of the agricultural drainage from the Delta appears to be the second most contaminated sampling location.

Another factor that may be responsible for the accumulation of pesticides in the Damietta branch is that its flow to the Mediterranean sea is stopped by a dam located at Damietta City (approximately 20 kilometers before the river exits to the Mediterranean sea). The dead-ended river (up stream of the dam) is the only source of drinking water for the city of

Table (4): Comparison between the concentrations of pesticides detected at all sampling locations and the lifetime health advisory level.

Compound	Mean conc. (µg/L)	Median conc. (µg/L)	Approximate lifetime health advisory level (µg/L)
Alachlor	199.6	189.8	0.4
Metolachlor	10.4	11.3	100
Pendimethalin	2.2	1.8	NA
Atrazine	4.1	3.2	3
Malathion	4.2	3.7	NA
Parathion	1.2	1.4	NA
DE-Atrazine	1.2	1.5	NA
Metribuzin	26.4	29.0	200
DIP-Atrazine	0.7	0.8	NA
Cyanazine	1.2	1.2	10
Trifluralin	0.1	0.1	2
Propachlor	0.5	0.5	90

Damietta, population approximately 904,000. Consequently the river water arriving at Damietta becomes highly concentrated with biological and chemical pollutants. In addition, this highly polluted part of the river at Damietta has been extensively used for fish farming, using isolated sections and cages, which adds to the pollution load from fish feed and farming activities. Frequent massive fish kills, in the Damietta branch as well as fish farms that use river or Manzala Lake water.

The mean and median concentrations of pesticides detected at sampling locations are listed in Table (4). Similar values were obtained for the mean and median concentrations at the four locations, indicating limited variability concentrations detected at the four locations. The mean concentration for alachlor (199.6 ppb) was approximately 500 times its health advisory level (0.5 ppb) recommended by the United States Environmental Agency. For atrazine, the mean concentration (4.1 ppb), or the median (3.2 ppb), were just above its health advisory level. When adding the mean concentration of atrazine metabolites (DE and DIP atrazine) to the mean concentration of the parent compound, the total atrazines will become approximately twice the health advisory level for atrazine. However, there is no health advisory level set for atrazine metabolites. The mean or median concentrations for other pesticides with listed health advisor level (metolachlor, metribuzin, cyanazine, trifluralin, and propachlor) were below that level. The health advisory for pendimethalin, malathion, parathion, and atrazine metabolites are not available. The River Nile at Cairo and its two branches, Damietta and Rosetta, constitute the primary drinking water source for the major part of the population in Egypt. This vital water resource is known to receive large portions of the drainage from heavy agricultural activities along the Nile in upper Egypt, and from the Delta surrounding its Damietta and Rosetta branches in lower (north) Egypt. The rest of the agricultural drainage in the delta is lifted mechanically into the

Manzala Lake at Damietta. In addition to being a water resource, both the River Nile and Manzala Lake represent a major fish supply for the rural population in the Delta. For Damietta Governorate, fish is the main source of protein and constitutes the major part of the urban and rural population. Thus the health advisory levels set for drinking water might significantly exceeded when combined with pesticide residue levels from fish and agricultural products. Therefore, it is important to monitor and eventually control the amount of pollutants in these vital and limited water and food sources.

CONCLUSION

The accumulation of pesticides in the river Nile and its major branches (Damietta and Rosetta), represent a major public health hazard to the Egyptian population which is concentrated at Cairo and thorough the Nile Delta. The increased contamination in the Damietta branch is possibly due the high volume of agriculture drainage discharged into the river as well as the lack of flow through the river at Damietta City. Although the pesticides detected have been widely used in Egypt over the last 20 years, to the best of our knowledge this is the first study to report the detection of those pesticides in surface water in Egypt. This is certainly due to the lack of analytical capabilities and effective monitoring program. There is also the need for routine monitoring programs and coordination effort to prevent discharging of agricultural drainage into the Nile River.

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