INVESTIGATION ON THE LEVEL AND MOVEMENT OF MERCURY CONTAMINANTS AROUND STORAGE AREAS AND FOOD PROCESSING FACTORIES IN HASSAHESA TOWN, SUDAN

By

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A

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DEDICATION

To my father "Ahmed", mother (Zeinab)
and uncle (Abdelwahab) with great
respect and love

CONTENTS

Page

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ABSTRACT

A total of 62 surface soil samples were taken from various sites in Hassahesa town and analyzed for total mercury level using X-ray fluorescence spectrometer.

Sites were chosen to represent the soil of pesticide store, food processing factories (involved in using contaminated seeds as fuel), near stock of redundant cotton seeds and neighbouring cotton fields. Control soils were sampled from similar soil type south Hassahesa town.

The results indicated an elevated mercury level in all samples analyzed far exceeding the background and normal range for mercury in soil. The highest level of contamination was found in soil of the pesticide store (51 ppm), followed by the two food factories (24 ppm and 19 ppm), stock of redundant cotton seeds (15 ppm) and cotton fields (14 ppm). Horizontal movement of mercury contaminants at various rates from foci of areas selected was noticed, wind direction and/or topography apparently had some role in this movement.

The level of total mercury in the control soil was exceeding the background and normal range for mercury in soils reported from other places. Various aspects of levels of mercury contamination, their movements, transportation and toxicological impacts on various forms of life were discussed.

LIST OF FIGURES

LIST OF TABLES

Table Page

LIST OF PLATES

CHAPTER ONE

INTRODUCTION

Organomercurials appeared as fungicides in the beginning of 1915. They have the advantage of reduced phytotoxicity, but with a wide range of fungicidal and bactericidal action. They functioned as a eradicants and offered an easy method of protecting seeds and seedling from seed and soil borne pathogens. Currently, organomercury compounds are widely used in world agriculture together with less amount of inorganic mercury compounds (Mcewen and Stephenson, 1979). The three forms of organomercurial employed in pesticide formulations are the alkyls, aryls and the alkoxyalkyls (Gayathri and Krishnamurthy, 1985).

The use of mercurials in agriculture and industry has been recognized as the major source of mercury pollution in the environment. This calls for thorough toxicity and mutagenicity investigation of the various forms of mercury that get into the ecosystem (Gayathri and Krishnamurthy, 1985). Hazards caused by organomercurials are well known. Many mercurials are highly volatile, irritant to skin and respiratory tract and toxic by oral, dermal or inhalation exposure. They accumulate in the living tissues and may lead later to kidney damage (Mcewen and Stephenson, 1979). Their use as fungicides in seed dressing caused many cases of poisoning episodes in Iraq, Guatemala, Pakistan, Sweden, Holland ... etc (Cook, 1977). Currently, several countries including the United States banned the use of organomercurial (Ware, 1983).

In Sudan, seed dressing with mercury fungicides started in 1930's (Saad, 1975). Later (in 1980), dressing of cotton seeds by Agrosan (phenyl mercury acetate) mixed with heptachlor or lindane was started. The banning of such compounds coupled with poor management practices has led to accumulation of huge quantities of not viable treated cotton seeds which were kept in open poorly fenced premises of cotton ginneries. Obsolete mercury compounds in Sudan were estimated at 975 kg and kept under similar conditions (Eltegani, 1998).

The accumulation of not viable treated cotton seeds led the authorities in Gezira scheme to sell significant amounts of treated seed as fuel to local food factories in Hassahesa town. Since these seeds were treated with organomercury compounds, then mercury vapour, mercury oxides or other toxic oxides like Co, SO_2 may emanate from such factories and cause severe deterioration in environmental quality there.

This situation initiated our interest to investigate the level and movement of mercury compounds around storage areas and involved food factories in Hassahesa town. Such study provides basic data necessary for any future toxicological or environmental risk assessment programme and possible corrective measures to the deteriorated environment.

The specific objective of the study can be summarized as:-

1) To determine the level of mercury contaminants over the foci of affected sites (stores, food factories).
2) To study their horizontal movement from the affected sites.

CHAPTER TWO

LITERATURE REVIEW

2.1 Heavy metals as fungicides

Among the heavy metals, only the compounds of copper and mercury have been used widely as fungicide although silver is the most toxic metal cation to fungi. The relative toxicity of various metals has been related to their position in the periodic table as Ag>Hg>Cu>Cd>Cr>Ni>Pb>Co>Zn>Fe>Ca (Cremlyn, 1978).

2.2 Mercury fungicides

2.2.1 Historical background

The use of mercurials as fungicides predated the twentieth century. Their first use was in seed-treatments, specially of cereals to control covered smuts and other seed-borne fungi (Mcewen and Stephenson, 1979).

2.2.2 Classification and forms

Nene and Thapliyal (1979) classified the mercurial fungicides into two types:inorganic (e.g: mercuric chloride, mercurous chloride) and organic (e.g: ethyl mercury chloride, phenyl mercury acetate ... etc).

From the toxicological and environmental point of view, the most useful and commonly accepted classification is done by Goldwater and Stopford (1977) who classified mercurials as:-

- Metallic mercury viz; liquid and vapour.
- Inorganic salts, such as sulphides, chlorides, nitrates and oxides.
- Alkyl compounds, such as those containing ethyl and methyl radial.
- Alkoxy alkyl compounds, usually of complex nature.
- Aryl compounds, such as the phenyl mercurials.

2.2.3 Organomercurials

The organomercurial include a wide variety of products with the general formula RHgX, where (R) represents an aryl, aryloxy, alkyl, alkoxy ethyl grouping and (X) is an anionic moiety such as chloride, acetate, hydroxy or other structures (Mcewen and Stephenson, 1979).

2.2.3.1 Development of organomercurials

In the early years, both mercuric and mercurous chlorides were used. Both are phytotoxic and therefore their use as fungicide was limited to seed or soil treatments for the control of diseases such as club root of cabbage and scab of potatoes (Mcewen and Stephenson, 1979).

The development of organomercurials resulted from the effort to find less toxic substitutes to inorganic mercurials (Nene and Thapliyal, 1979). They had reduced phytotoxicity with wide spectrum of activity against variety of bacterial and fungal organisms (Mcewen and Stephenson, 1979).

The first commercial product was Uspulum which contain 18.8% mercury in the form of chlorophenol mercury (probably $Cl(OH)C_6H_3$ $Hg\text{-}OSO_3$ Na). This compound was introduced in 1915 by the Bayer Company of Germany. Subsequently Germisan was introduced around 1920 (Nene and Thapliyal, 1979).

2.2.3.2 Uses of organomercurials

Mercury, especially organic mercuries functioned as an eradicant to kill spores and prevent development of dormant mycelia. It was learned that mercurials were broadly toxic to fungi and bacteria (Mcewen and Stephenson, 1979). Organomercurials, in particular, have an exceeding wide and high fungicidal activity and are mostly used for seed dressing owing to their wide activity and very low application rates (Kramer, 1983).

Seed treatments offered an easy method of protecting seeds and seedling, not only from seed-borne pathogens but also from soil borne pathogens (Mcewen and Stephenson, 1979). The use of mercury for treating cereal and sugar beet seed began in the 1920's. Fusarium, Helminthosporium, Ustilago and Septoria are among the principal cereal disease (Nelson et al., 1971).

Among potent organomercurial in current use is phenyl mercury acetate (PMA). This compound was described by Worthing (1968) as a powerful eradicant fungicide mainly used for treatment of cereal seeds, often in combination with insecticides or other fungicides.

The commercial preparations recommended for dry seed treatment by mercurials should contain 1% metallic mercury (Nene and Thapliya, 1979), or about 1.5% mercury (Nelson *et al.*, 1971).

2.2.3.3 Mode of action of organomercurials

In contrast to the inorganic metal salts, organometallic fungicides are mostly volatile and thus act via the gas phase (Kramer, 1983).

Lindstrom (1958) studied the action of mercurials and concluded that coverage occurs in two steps; distribution on the seed in the treating equipment and redistribution by evaporation and resorption of the compound. In the later process, the chemical tends to leave locations where it is abundant and be resorbed where it is scarce, so that in short time uniform coverage is achieved. Lindstrom believed that two kinetic processes are involved in the movement of mercurials; namely evaporation and diffusion rates and of the two he considered diffusion to be the slower or the rate controlling step.

According to Metcalf (1971), the stability of the organomercurial and the rate at which Hg^{2+}ion is released, is determine by the alkyl or aryl portion of the molecule which provide the lipophillic character to the molecule and thus aids in penetration into the cuticular layer. It is generally accepted that the site of fungicidal action of mercury, either as vapour or ion, is at the sulfhydryl (-SH) groups of the susceptible enzyme (Nene and Thapliyal, 1979). Mercury compounds were non-specific inhibitors for all enzymes. The metal reacts with thiol groups in the cell and form more stable complexes with the -SH compounds (Lindstrom, 1958).

2.2.3.4 Fate of organomercurial in the soil environment

Generally, the distribution and fate of pesticides are determined by a number of variables which include the nature of pesticide and environmental factors. Figure (1) summaries the processes influencing the behavior and fate of pesticides in the soil environment (Mcewen and Stephenson, 1979).

The degradation of organomercury fungicides in the soil has been investigated by Kimura and Miller (1964). They detected both metallic mercury and volatile organomercury compounds in the air above soil which had been treated with ethyl or phenyl mercury acetate or methyl mercury derivatives. They concluded that the degradation process was mainly microbial, although a large proportion of the mercury compounds persisted intact in the soil over periods of 30-50 days. Earlier, Booer (1944) confirmed in his studies that organomercury salts decomposed rapidly into metallic mercury in soil by reduction and disproportionation reactions via mercury (I) chloride.

Recently, however, the microbial degradation of phenyl mercury acetate has been the subject of various studies. Using the Hg^{203}-labelled compound, Tonomura *et al*. (1968) found that mercury-resistant organism K62, isolated from soil, seemed

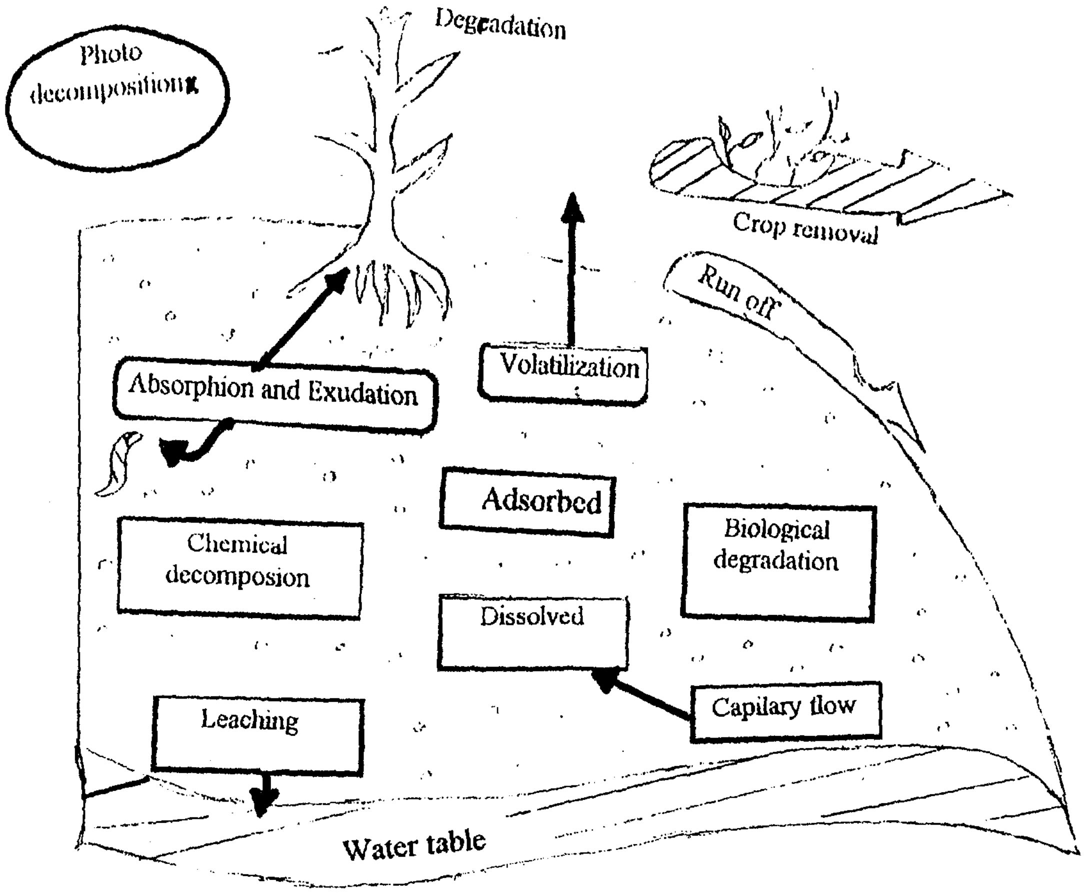


Fig.1 :

Processes influencing the behavior and fate of pesticide in the soil environment

able to bring about a conversion to a substance more volatile than phenyl mercury acetate. Subsequent efforts by Furakawa *et al.* (1969) using Hg^{203} or C^{14}-labelled compound revealed that some 70% of Hg^{203} or 80% of C^{14} disappeared in two hours on an aerobic incubation with washed cells previously grown in the presence of 20 ppm of phenyl mercury acetate. Fission of the carbon-mercury bond yielded metallic mercury together with benzene detected by Gas liquid chromatography (GLC), no benzene was detected in an abiotic control experiment.

A further advance study was made by Matsumura *et al.* (1971) who incubated Hg^{203}-labelled phenyl mercury acetate at 30°C for 10 days with culture of some 35 microorganisms isolated from natural lake bottom sediments or soils. The major metabolite produced by all of the microbial culture was dipheny mercury, but a second metabolite was not identified and there was no evidence of any direct conversion of phenyl mercury to methyl mercury ($MeHg^{+}$). In contrast, Cremlyn (1978) insured the conversion of metallic mercury, inorganic Hg^{+2} ions and organic mercury compounds in the environment into methyl mercury which can then be taken up by living organisms. It was thought that the methyl mercury found in the food chain of fishes originated primarily from phenyl mercury acetate extensively used as fungicide (Westoo, 1966; 1967). It is not known where and how the transformation into methyl mercury occurs (Kivimae *et al.*, 1969) and the direct conversion of phenyl mercury to methyl mercury is apparently not a common process in microorganisms. At least one species of microorganisms is known (Tonomura *et al.*, 1968) to be capable of direct conversion of phenyl mercury to metallic mercury, a process which could eventually lead to the formation of methyl mercury through the conversion of Hg° to CH_3Hg^+ (Matsumura *et al.*, 1971).

As long ago as the sixteen century, it was suggested that there was a "Mercury cycle" whereby the metal circulate through the lithosphere, atmosphere, hydrosphere and biosphere. This cycle has recently portrayed in various ways (Fig. 2) and it is now been demonstrated that methylation can occur in nature only under very specific and limited conditions of mercury concentration, acidity, temperature and bacterial species. Not all mercurials are converted to methyl forms. Phenyl mercury acetate, for example, is converted to diphenyl mercury. Demethylation also can occur preventing excessive or dangerous accumulation of methyl mercury (Fig. 2). The cycle of mercury has recently shown to be quite complicated. Microorganisms are able to reduce mercury (II) salts to metallic mercury and/or methyl mercury derivatives or even to the highly toxic dimethyl mercury (Goldwater and Stopford, 1977).

2.2.3.5 Residue distribution

Many synthetic pesticides are exceptionally persistent. They remain in soil; move with water; absorbed by plants; accumulate in animal fat and are transferred to milk and eggs

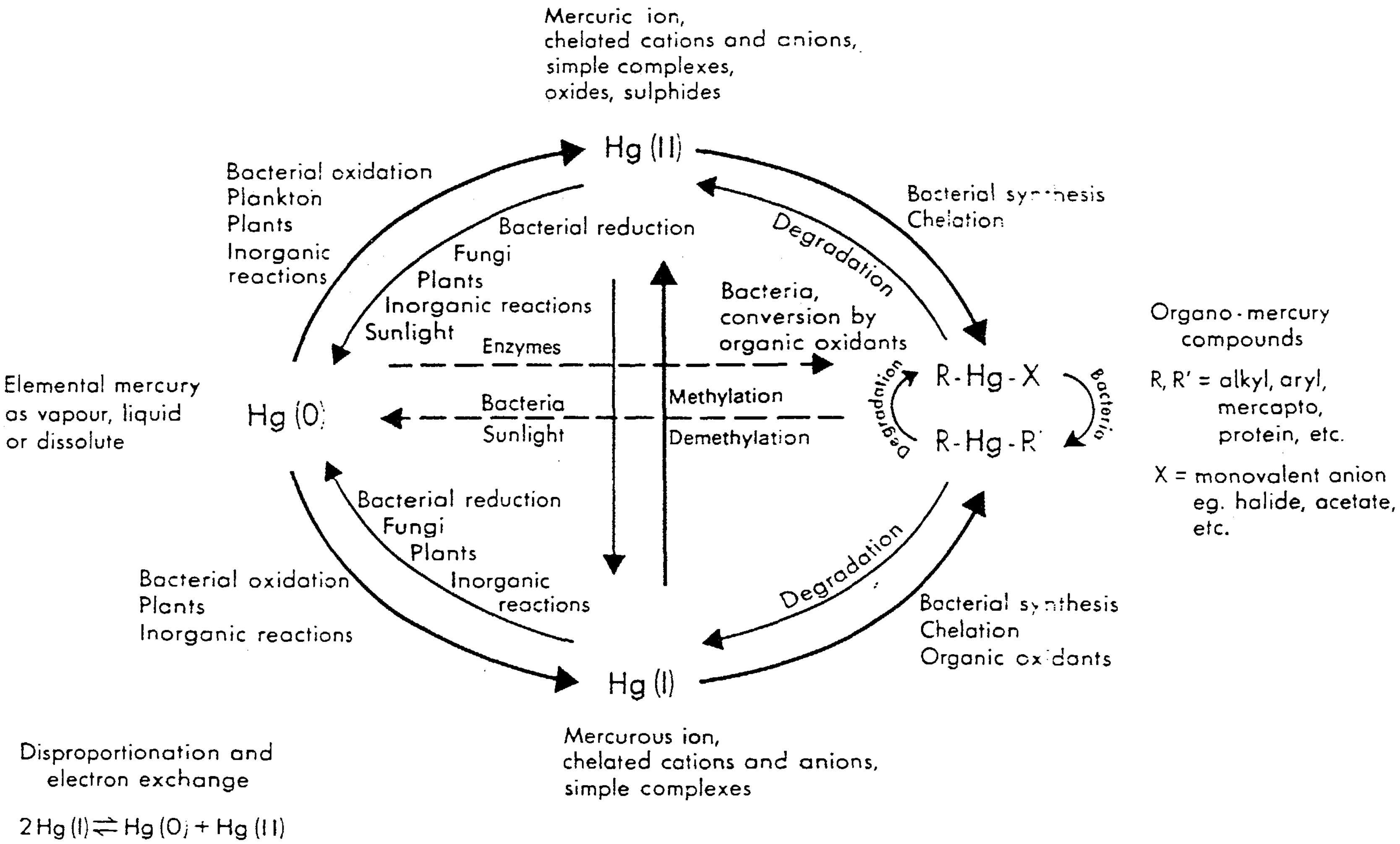


Figure 2. Cycle of mercury interconversions in nature

From Goldwater and Stopford (1977)

(Chichester, 1965). Considerable evaporation of these chemicals or their toxic derivatives may occur, producing atmospheric pollution (Elbeit *et al.*, 1977).

The overall concentration of mercury in, on and around the earth is constant (Table 1). Man ingenuity have synthesized hundreds of mercury compounds far more than those resulted from natural conversion (Goldwater and Stopford, 1977). Highly crossive and toxic residues of pesticides and minning industries have been accumulated in the neighbourhood of many parts of the world without due consideration of their devastating impacts on the local environment or danger to human health (Tolba and Elkholy, 1972-1992).

2.2.3.5.1 Organomercury residues in soil

Furutani and Osajima (1965a and b) reported the mercury content of a well-drained paddy field soil where organomercury compounds had been applied as dust to be 0.33 ppm and 1.36 ppm for an ill-drained soil, while Stock and Cucuel (1934) reported up to 1.7 ppm mercury in garden soil. Higher value was detected by Pickard *et al.* (1962) who found 2.6 ppm of mercury in soil treated once with yellow mercuric oxide to control potatoes root eelworm.

The natural mercury content of soil was reported by a number of scientists. Martin (1963) reported the natural mercury content of some English soil as between 0.01 and 0.06 ppm, while Anderson (1967) reported an average of 0.01 to 0.9 ppm in some 200 analyses of Swedish soils and lower values of

Table 1: Normal ranges for total mercury in the environment*

Lithosphere	0.00 - 10.0 mg/kg
Hydrosphere	0.01 - 6.00 μg/L
Atmosphere	0.005 - 1.0 μg/m^3
Food (fresh weight)	0.01 - 1.5 mg/kg
Human urine	1.0 - 25.0 μg/L
Human blood	1.0 - 50.0 μg/L
Human hair	1.0 - 5.0 mg/kg

* From Goldwater and Stopford (1977).

0.05 ppm were reported in French and Sudan soils. The actual values vary appreciably with the locality where the sample was taken. Table (2) shows the content of mercury in soils.

The average mercury content of the earth's crust was reported as 0.5 ppm while the background level of mercury in soil ranges from 0.1 to 2.0 ppm (Saha, 1972).

2.2.3.5.2 Organomercury residues in water

Concern about mercury in drinking water is of recent origin. There was no mention of this metal in the widely used US public health service drinking water standards of 1962.

Jervis and co-workers (1970) reported on the mercury content of several fresh water bodies; fishes; meat; wheat and milk powder. Several of the reported values exceed 200 ppb.

The Soviet Union was the first nation to set standards for mercury in drinking water in 1950, as 0.005 mg/L (5 ppb) for mercury or mercuric ions. While the World Health Organization (1970) recommended a tentative upper limit of 1.0 μg/L. Saha (1972) stated the background level of mercury in river water as 0.1 ppb, 0.01 to 0.05 ppb in ground water, 0.03 ppb in sea water and 0.2 ppb in rain water.

2.2.3.5.3 Organomercury residues in air

Nelson and his co-workers (1971) thought that much of the atmospheric mercury is in the form of vaporized dimethyl mercury and reaches the air primarily through evaporation from soil or water. However, both monomethyl and metallic mercury

Table 2: Content of mercury in soils*

Soil type	Concentration (%)
Humic horizon of forest soils	3.0 - $8.1\text{x}10^{-6}$
Forest soils	10.0 - $29.0\text{x}10^{-6}$
Cultivated soils	3.0 - $7.0\text{x}10^{-6}$
Clay soils	3.6-$3.4\text{x}10^{-6}$
Sand soils	0.1-$2.9\text{x}10^{-6}$

* From Stock and Cucuel (1934).

are also volatile and nothing is known about stability of organic mercury in the atmosphere.

Very little is known about either the quantitative or qualitative aspects of mercury in the atmosphere. The Swedish literature cites work done in Germany in early 1930's by Stock and Cucuel (1934) who estimated an average value of total mercury in air as 0.02 $\mu g/m^3$. While Cholak (1952) gave values ranging from 0.03-0.2 $\mu g/m^3$. More sensitive measurement of mercury vapour was done in Sanfransisco bay area (Williston, 1968), were winter values ranging from 0.001-0.025 $\mu g/m^3$ and summer values ranging from 0.002-0.50 $\mu g/m^3$ were recorded.

The background level of mercury in the atmosphere, under normal conditions as set by Goldwater (1971) and Saha (1972) should be less than one part per billion.

2.2.3.5.4 *Organomercury residues in food*

Humans get certain amount of mercury from food, water and air. Of these three sources, only food is of any consequence under ordinary circumstances (Goldwater and Stopford, 1977) [Fig. 3].

All food contains traces of natural mercury. Plant and animals tend to concentrate mercury and in some instances the levels of mercury in some animal tissue can be dangerous to human health (Saha, 1972). In aquatic environment, some fish at the top of the food chain, such as the pike, contain about 3000 times more mercury than the water (Hannerz, 1968).

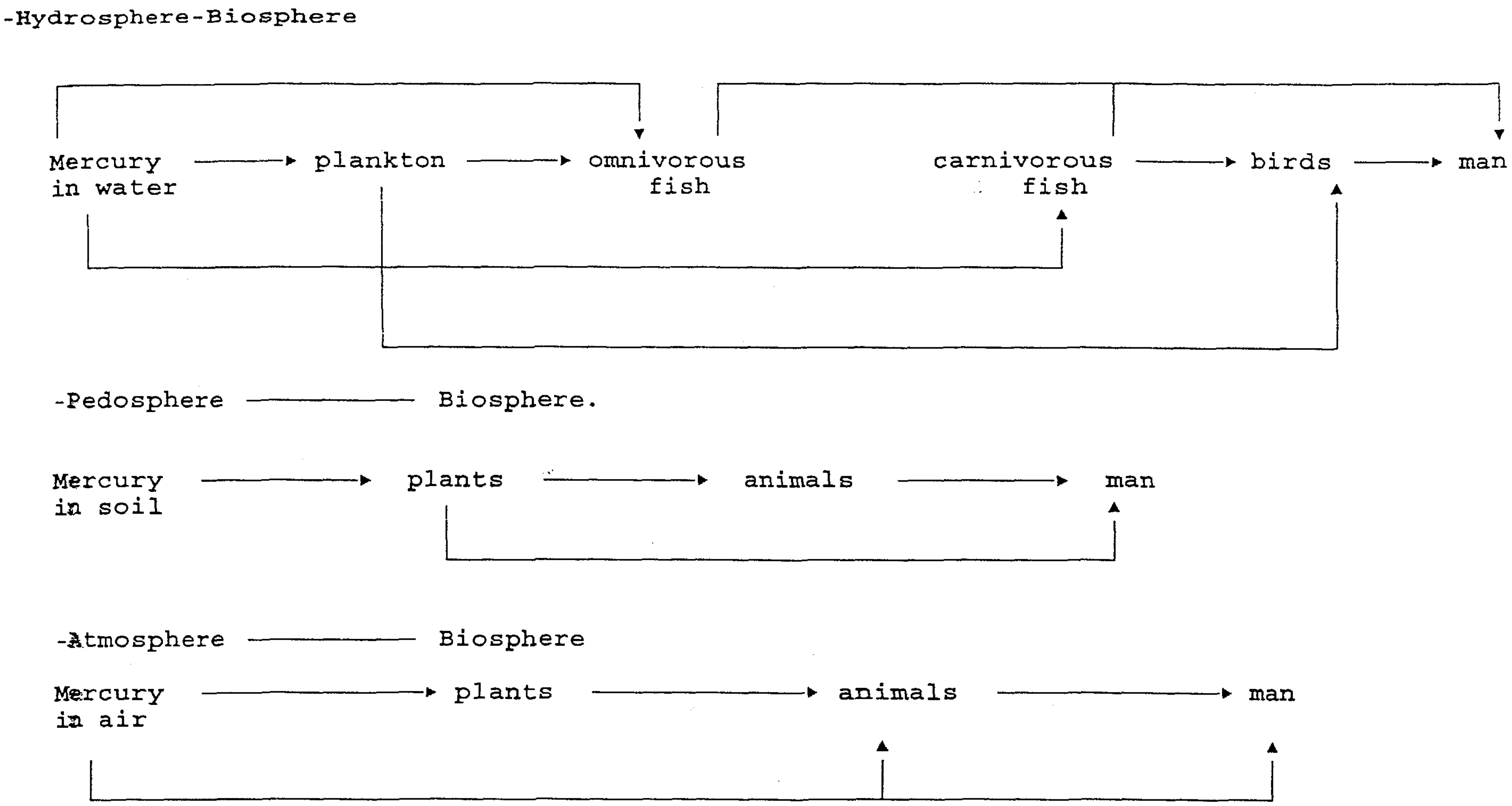


Fig. 3: Generalized food chains according to the geological survey of Canada (Goldwater and Stopford, 1977).

The contamination of crops with mercury used in agriculture has been reviewed by Smart (1968). He concluded that in some fruit, the background levels are normally 0.04 ppm of mercury or below; in tomatoes up to 0.02 ppm; in potatoes up to 0.01 ppm; in wheat and barley up to 0.02 ppm and in eggs and meat up to 0.05 ppm. When a crop or foodstuff is treated in accordance with agricultural practices, residues of mercury are generally not greater than the following: apples 0.1 ppm; tomatoes 0.1 ppm; potatoes 0.05 ppm; wheat and barley 0.02 ppm and eggs and meat 0.1 ppm. It can be seen that the agricultural use of mercury compounds could increase mercury levels in some foods and grains grown from treated seeds (Saha, 1972; James *et al.*, 1971). Table 3 shows mercury residues in various types of food.

The mercury content of Sweden eggs was higher than that in eggs from European countries and this is due to the seed dressing with mercury compounds in Sweden (Westoo, 1969).

FAO/WHO in Rome 1963 set tolerable levels for mercury in certain types of food as 0.05 ppm total mercury. This standard specifically excluded drinking water; fish and shell-fish. In 1968, they have discussed the question of organomercurial compounds in food and suggested a tolerance limits of 0.02-0.05 ppm. Table 4 shows standards of mercury in foods in certain countries.

Table 3: Mercury residues in food (ppm)

Food	Stock and Cucel (1934) Germany	Gibbs *et al.* (1941) U.S.A	Goldwater (1964) U.S.A
Meats	0.001-0.067	0.0008-0.044	0.001-0.15
Fish	0.02-0.18	0.0016-0.014	0.0-0.06
Fresh vegetables	0.002-0.044	0	0.0-0.06
Fresh milk	0.0006-0.004	0.003-0.007	0.008
Grains	0.02-0.036	0.002-0.006	0.002-0.025
Fresh fruits	0.004-0.01	-	0.004-0.03
Eggs	0.002	0	-
Beer	0.00007-0.0014	-	0.004

Table 4: Standards of mercury in foods*

Country	Type of food	Tolerance (ppm)
S. Australia	All	0.01
W. Australia	All	0.01
Brazil	All	0.05
Canada	Fish	0.50
Denmark	All	0.05
Germany	Fruits, vegetables	"Zero"
Japan	Fish	1.00
Newzealnd	Fruits, vegetables	0.05
Sweden	Fish	1.00
	Other foods	0.05
U.S.A	Fruit, vegetables	"Zero"
	Fish	0.50

* From Goldwater and Stopford (1977).

2.2.3.5.5 Organomercury residues in animals tissues

Borg *et al.* (1966) found very high levels of mercury in the livers and kidneys of birds found dead in Swedish countryside (table 5) and lower levels (1-53 ppm) in live birds. The source of mercury was attributed to seed-dressing used in Swedish agriculture (Tejning, 1967). Studies in Canada (Gurba, 1970; Fimreite *et al.*, 1970) had shown mercury contamination of seed-eating birds can be transferred to their predators.

2.2.3.6 Toxicological and physiological aspects of organomercurial compounds

The various chemical and physical states of mercury have been known to pose serious threats to environmental health (Goldwater, 1972; Evans *et al.*, 1975; EPA, 1984). Mercury, like most heavy metals was known to be persistent in environmental and biological systems, especially the central and peripheral nervous system (Watanable, 1969; Oehme, 1978a and b; Hamilton, 1981).

Extensive studies of human exposure to phenyl mercurials conducted at Columbia University led to the conclusion that phenyl mercurials may be absorbed through the intact skin or mucus membranes, but relatively high concentration must be applied before measurable amounts will be absorbed in this way.

Mercury solution is irritant to skin and respiratory tract, caused second-degree of chemical burn and toxic by oral, dermal or inhalation exposures. Toxicity to man is

Table 5: Mercury residues in some types of Swedish birds

Bird	Concentration (ppm)
Pheasants and partridge	28-140
Pigeons	8-45
Corvine birds	29-110
Finches	11-136
eagles; buzzard; hawks and falcons	6-100
Owls	4-27

expressed as nervousness and severe impairment of coordination (Goldwater and Stopford, 1977). Phenyl mercury acetate was shown to cause damage in the kidneys (Fitzhugh *et al.*, 1950).

The use of organomercurial fungicides in seed-dressing, especially for grain seed, has given rise to outbreak of poisoning, in Iraq, 1973 where more than 200 people died as a result of the consumption of home made bread prepared from dressed wheat. Similar occurrences has been reported from Guatemala and Pakistan (Goldwater and Stopford, 1977). Poisoning by consumption of treated seeds have been reported also in livestock, domestic and wild birds (Mcewen and Stephenson, 1979). Exposed birds may show toxic symptoms followed by laying of eggs with thin shell. If they hatch at all they may give rise to chicks with poor survival, therefore wildlife is put at risk (Cook, 1977).

Industrial effluent has been major source of mercurial pollution. But the increase in mercury contamination was attributed to the use of wide range of mercurial compounds in seed dressing (Briggs and Courtney, 1985).

2.2.3.7 ***Pesticides storage and misuse in Sudan***

The movement of imported pesticides from Port Sudan to the other areas is normally carried by railways, trucks or lorries. This transport system was recognized as inefficient and slow, therefore imported chemicals stay~~s~~ for long period in the open under poor storage conditions. Such conditions lead to deterioration of containers by the action of high humidity (ave. 55%) and worm condition (ave. 29°C) at Port Sudan. The rough handling of containers, other environmental factors such as wind, rain, temperature may contribute to further deterioration of the imported chemicals or containers (Saad, 1975).

Butrous (1990) studied the condition of storage facilities, quality and quantity of obsolete pesticides and their method of disposal. The study covered the Blue Nile area (Suki), Elrahad, New Halfa and Gezira scheme. He found that most of the visited sites have no proper pesticide stores and even the existing ones were of poor ventilation and not suitability located. He suggested that the visited sites needed construction and/or rehabilitation of pesticides stores.

The improper storage is one of the most serious drawback of pesticides use in Sudan which causes serious health problems among the local communities around pesticide stores.

There are two main reasons for the accumulation of obsolete pesticide in plant protection directorate (PPD) and agricultural corporation stores. First, the ban issued in 1980 (in Sudan) on the use of DDT and some other chemicals which were stocked in large quantities. Secondly, the general lack of environmental awareness among officials resulted in the underestimation of the problem so that the fund and facilities allocated for storage and prevention of accidents are too meagre to protect the people and environment (Abdellatif, 1995).

Considering, such huge quantities of obsolete pesticides (Table 6) one should expect serious contamination of the soil in places where these pesticides are stored which in itself requires some sort of treatment (Eltegani, 1998) [Plates 1, 2, 4 and 5].

2.2.3.8 Intoxicated cotton seeds

In November 1993 and during the work on Hassahesa pesticide grave yard by the Sudanese Development Agency team (SDA), a new problem, quite hazardous as the dumping of obsolete and banned pesticides emerged. It concerned with accumulation of intoxicated cotton seeds in many areas around the pesticide stores in Hassahesa and Maringan ginneries and near the dumping site (Plate 4 and 5).

The seeds were treated with Agrosan (phenyl mercury acetate) mixed with either heptachlor or lindane. The banning of such chemicals led to the accumulation of treated seeds which Lost their viability by either long storage and/or exposure to extreme weather condition.

This situation led The authorities of Gezira scheme to sell significant amount (20,000 sacs) of treated cotton seed as fuel to local food factories in Hassahesa town. later on (1995), the health inspector of Hassahesa province banned the burning of treated cotton seeds until he gets scientific advice . He asked the help of SDA, where a small committee consisting of an occupational health inspector, chemist and an environmentalist was formed, their plan was to find out the

Table 6: **List of obsolete chemicals in Sudan***

Type of chemicals	Quantity
Organophosphates	99.049 L
Chlorinated hydrocarbons	150345 L
Carbamates	2790 L
Pyrethroids	2590 L
Mixtures of chlorinated hydrocarbon and organophosphates	2864 L
Mixtures of pyrethroids and organophosphates	3900 L
Anticoagulants (Rodenticides)	31135 kg
Unidentifiable solid products	128100 kg
Unidentifiable liquid products	30930 kg
Urea	4650 kg
Mineral oil (olbolinum)	22400 L
Arsenate	10000 kg
Mercuric compounds	975 kg
Sulfur	400 kg
Sodium Cyanide	400 kg
Naphthalene anhydride	21000 L

* From Butrous *et al.* (1990).

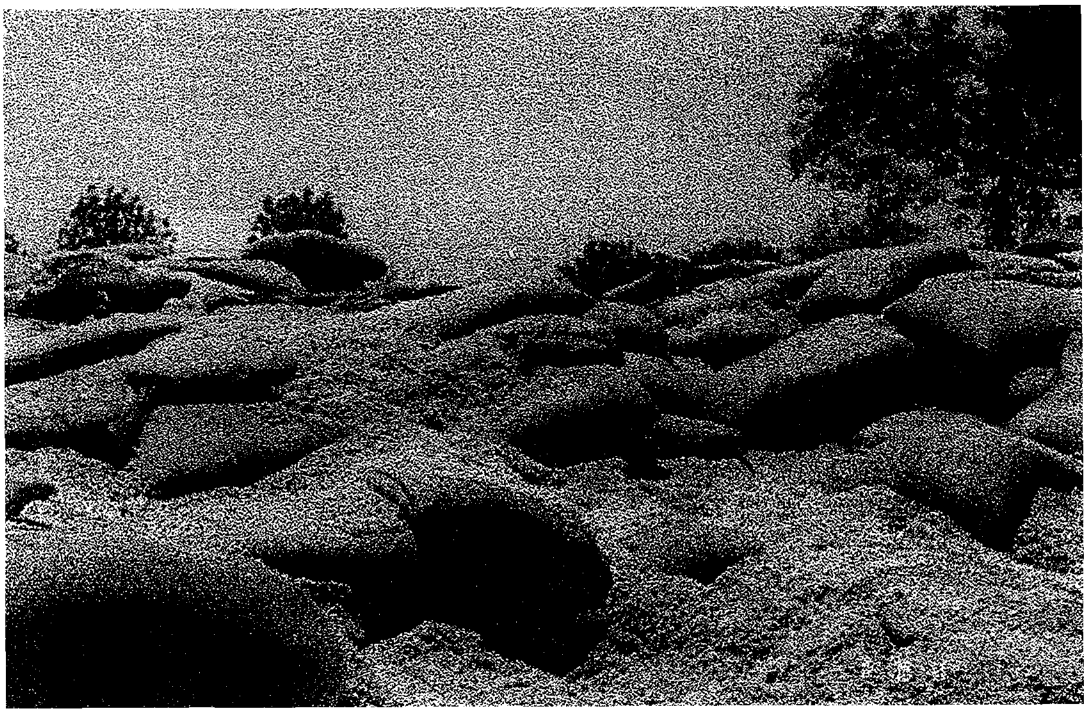


Plate (1): The status of treated and untreated stock of cotton seeds at maringan Ginneries

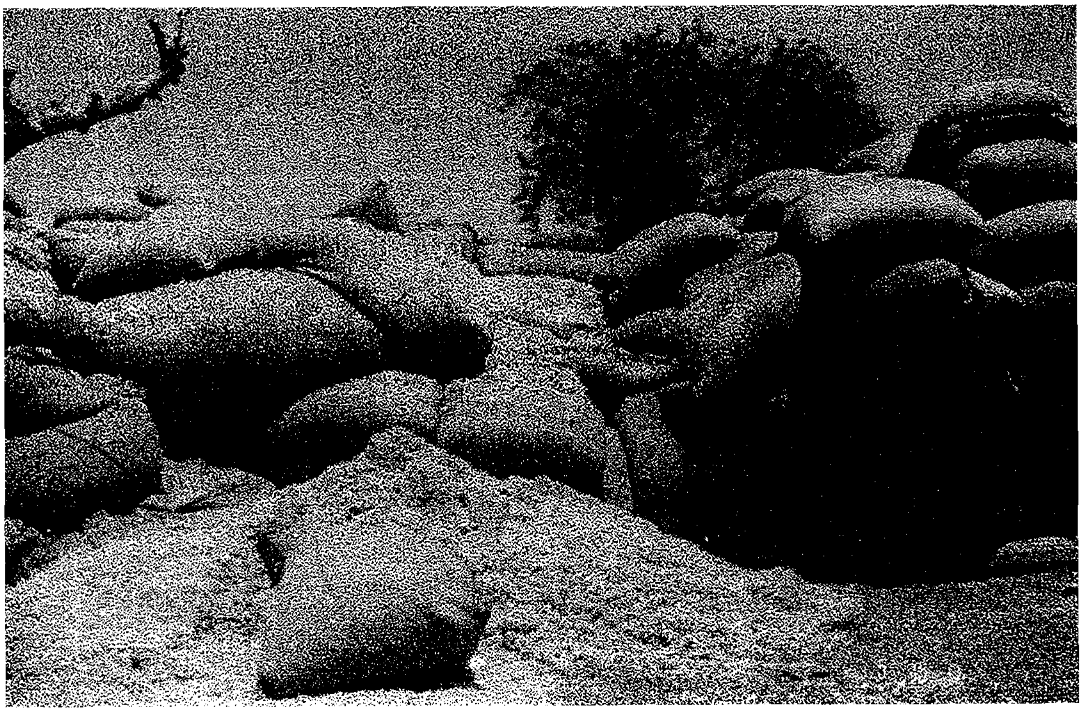


Plate 2 : obsolete stocks of cotton seed treated by different pesticides at maringan Ginneries

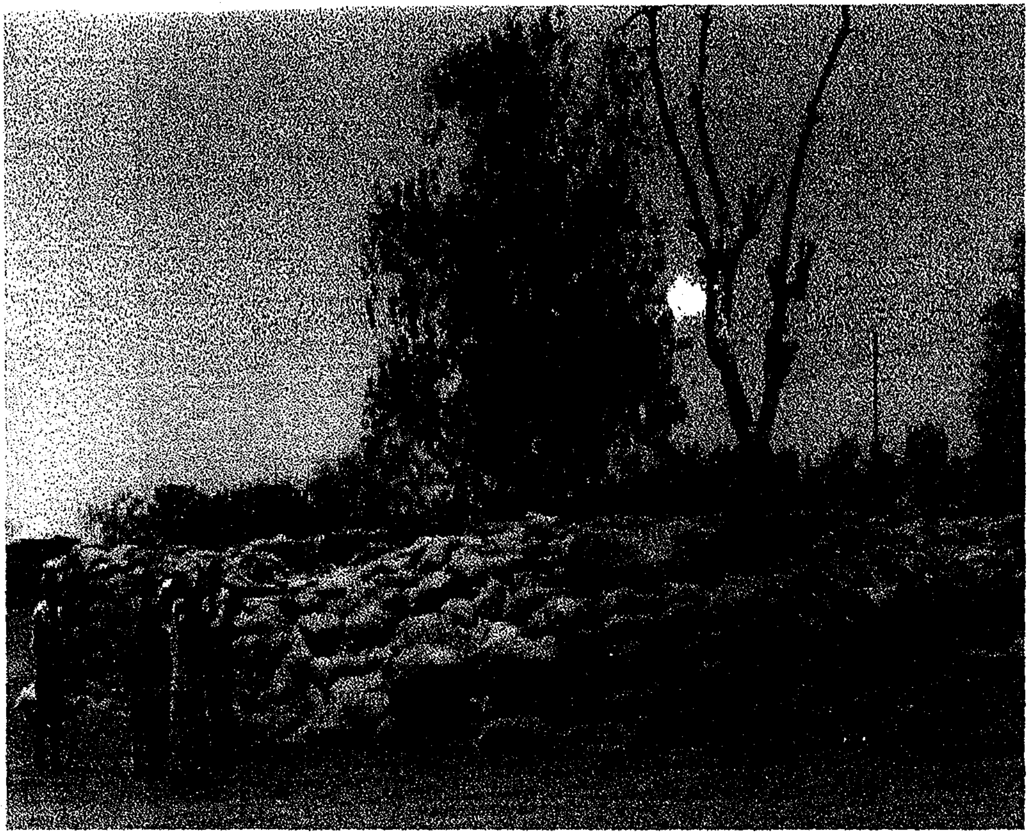


Plate (3) : The death of Neem tree due to the effects of treated cotton seeds

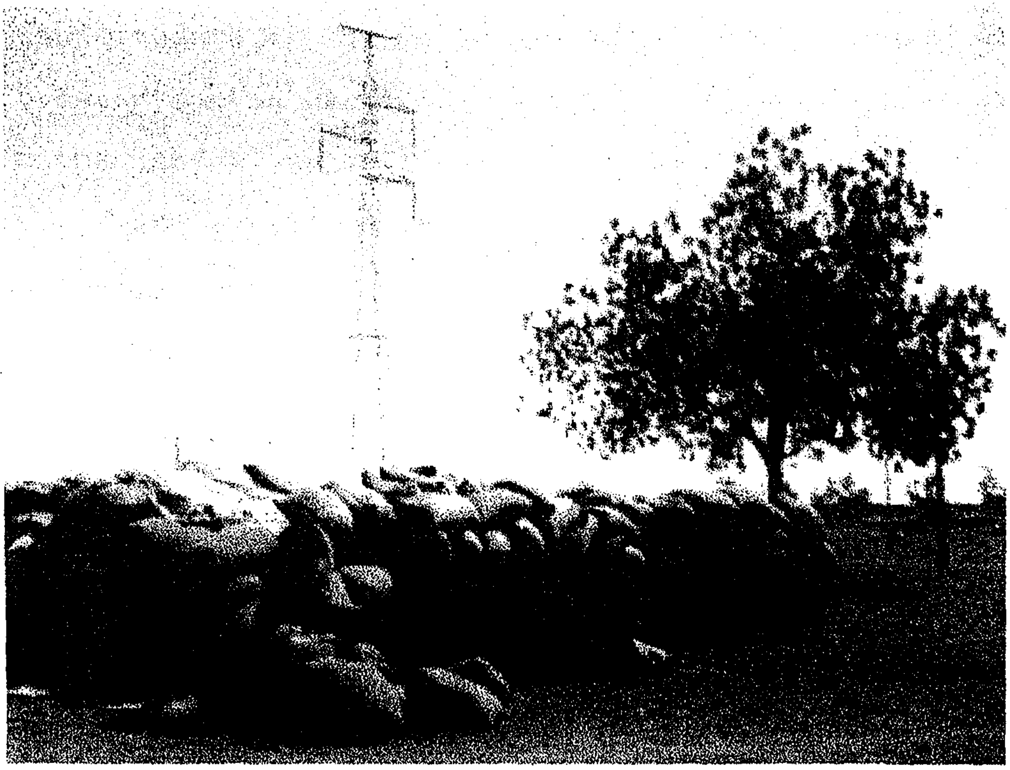
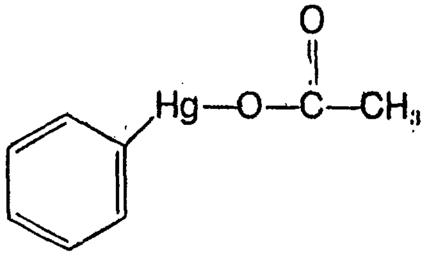


Plate (4) stock of cotton seed treated by A grosan /helptachlor at Hassahesa Genneries

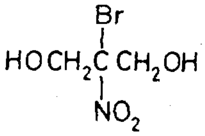
Plate (5) : stock of cotton seed treated by Agrosan/lindane at Hassahesa Ginneries constituents of the fumes and ashes of burn seeds. Samples of treated cotton seeds were taken for burning under lab conditions (Abdellatif, 1995).

Results indicated that part of mercury evolve as vapour while some remains on the ash. Large quantities of CO, CO_2, SO_2 and tolwin liberates (Elmaheina, 1995). This may indicate the possibility of air contamination by toxic mercury vapour emanating from the burning of treated cotton seeds in food factories. As mercury vapour (or oxides) are heavy they may quickely fall on the neighbouring areas of the burning sites and causes severe deterioration in the environmental quality there. Health of residence and domestic animals in the neighbourhood of these factories and pesticide stores is greatly endangered by such accident. Corrective measures to such contaminated environment is badly needed.

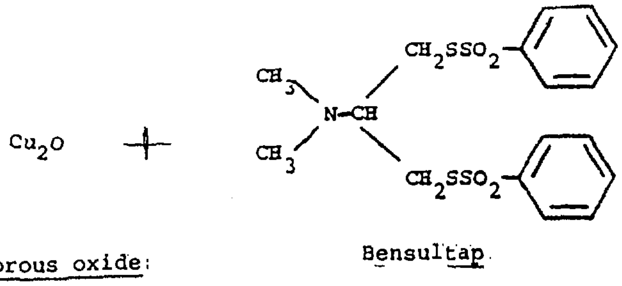
PMA



Agrosan
(phenylmercury acetate)

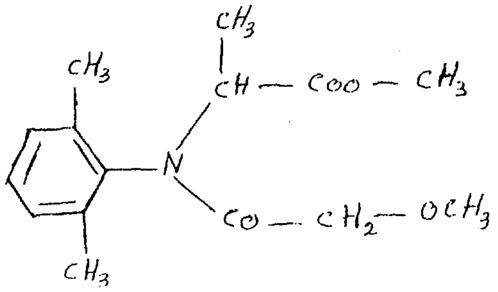

$\mathrm{HOCH_2\overset{Br}{\underset{NO_2}{C}}CH_2OH}$

Bronopol

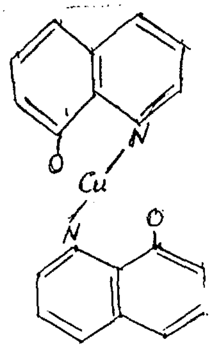

Cuprous oxide:

Bensultap.

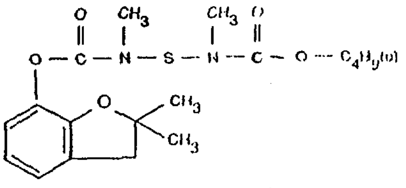
Supersan
(Cuprous oxide/bensultap)



Apron 200 Ls
(metalaxyl



Quinolate 150 plus
(Copper oxinate)



Promet 400 CS
(Furathiocarb)

Fig . 4 : Structural formula of fungicides mentioned in the study.

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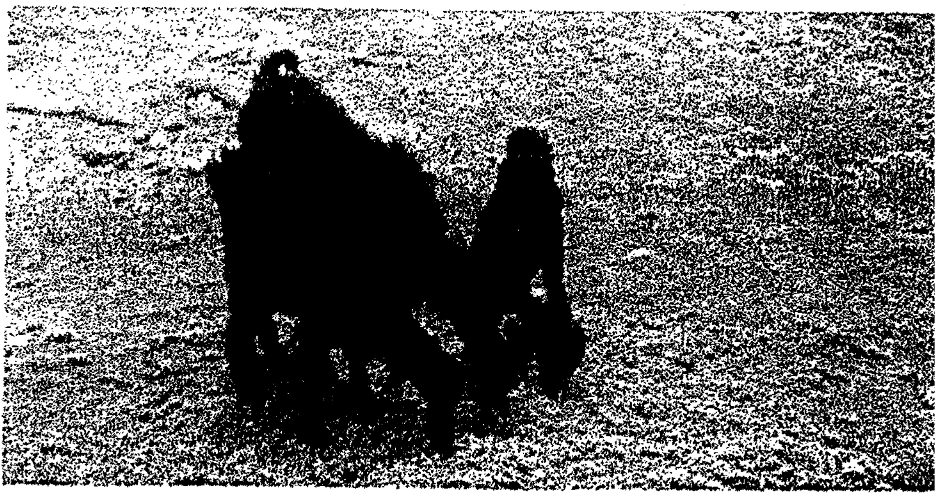

Plate (6) grazing animals all over the cotton Ginneries at Hassahesa

CHAPTER THREE

MATERIALS AND METHODS

3.1 Site description

Soil samples were collected from five locations within Hassahesa town, Gezira State, Sudan, for estimation of total mercury level. The first two sites, pesticide store and stock of treated cotton seeds, were located at the western part of Hassahesa town, near the pesticide dump pits and within the fence yard of Hassahesa cotton ginneries. The third and fourth sites were located at the food processing factories which engaged in using the intoxicated cotton seeds as fuel. The factories are located north of Elmazad compound, Hassahesa town. The fifth site is located at the cotton fields, west Gezira Main Canal near Wad Sulfab (Fig. 5a). Surrounding the first four selected sites lies six residential camps:-

1. Irrigation living compounds
2. Quarashi compounds
3. Kurmuk compounds
4. Campo compounds
5. Bank house compounds
6. Elmazad house compounds

Control soils were sampled from similar soil type at Arqueet area in the southern part of Hassahesa town. They are used for determination of background level of mercury in the soil of this area.

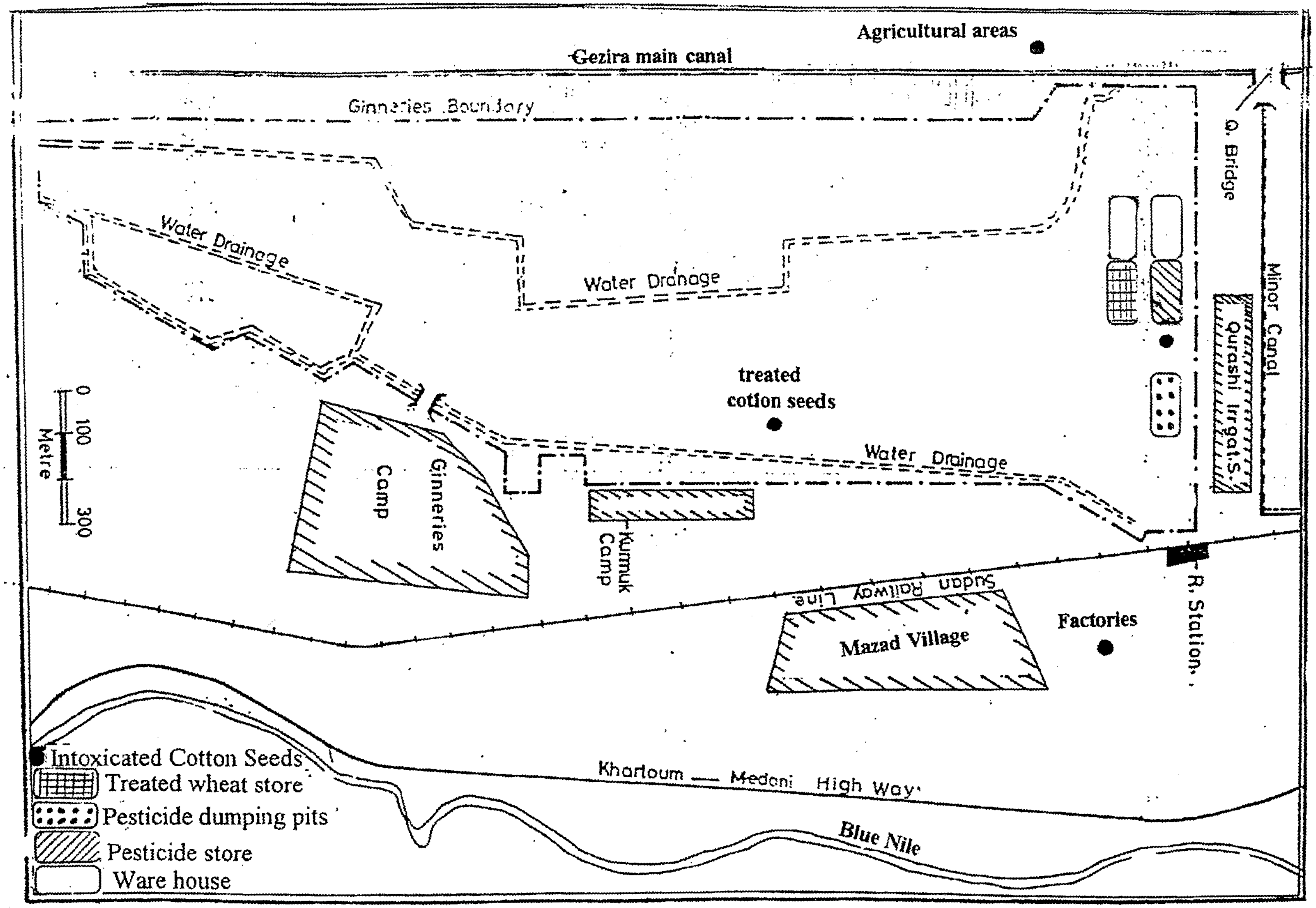

Fig. 5a; Location of study area (General)

3.2 Soil sampling

Surface soil samples (0-10 cm) were taken at variable distances (0, 50, and 100 m) and directions from the selected sites (Fig. 5b).

Specifically samples were distributed as follows:-

1] Four samples from, the centre of the pesticide store; 100 meters east; 100 meters north and 100 meters south.

2] Three samples were taken near the stock of treated cotton seeds and along the slope gradient from south to north. Two samples, 100 meters apart, were taken close the heap of treated cotton seeds. The third sample was taken 100 north the second sample and the seed sacs (Fig 5b).

3] Five samples were taken from, the centre of factory I yard, 100 meters east; 50 meters south; 100 meters south and 50 meters north.

4] Five samples were taken from the centre of factory II yard and 50 meters in the four directions.

5] Two samples were taken from neighbouring cotton fields at the west side of Gezira main canal at Wad Sulfab village.

6] Control soil was sampled from Arqweet area, south Hassahesa town.

Each site was sampled three times, soil samples were wrapped separately in polyethylene bags and bought to the X-ray lab, Physics Department, University of Khartoum for total mercury determination. Subsamples were taken from each sample and brought to soil lab, Soil Department, Faculty of Agricul-

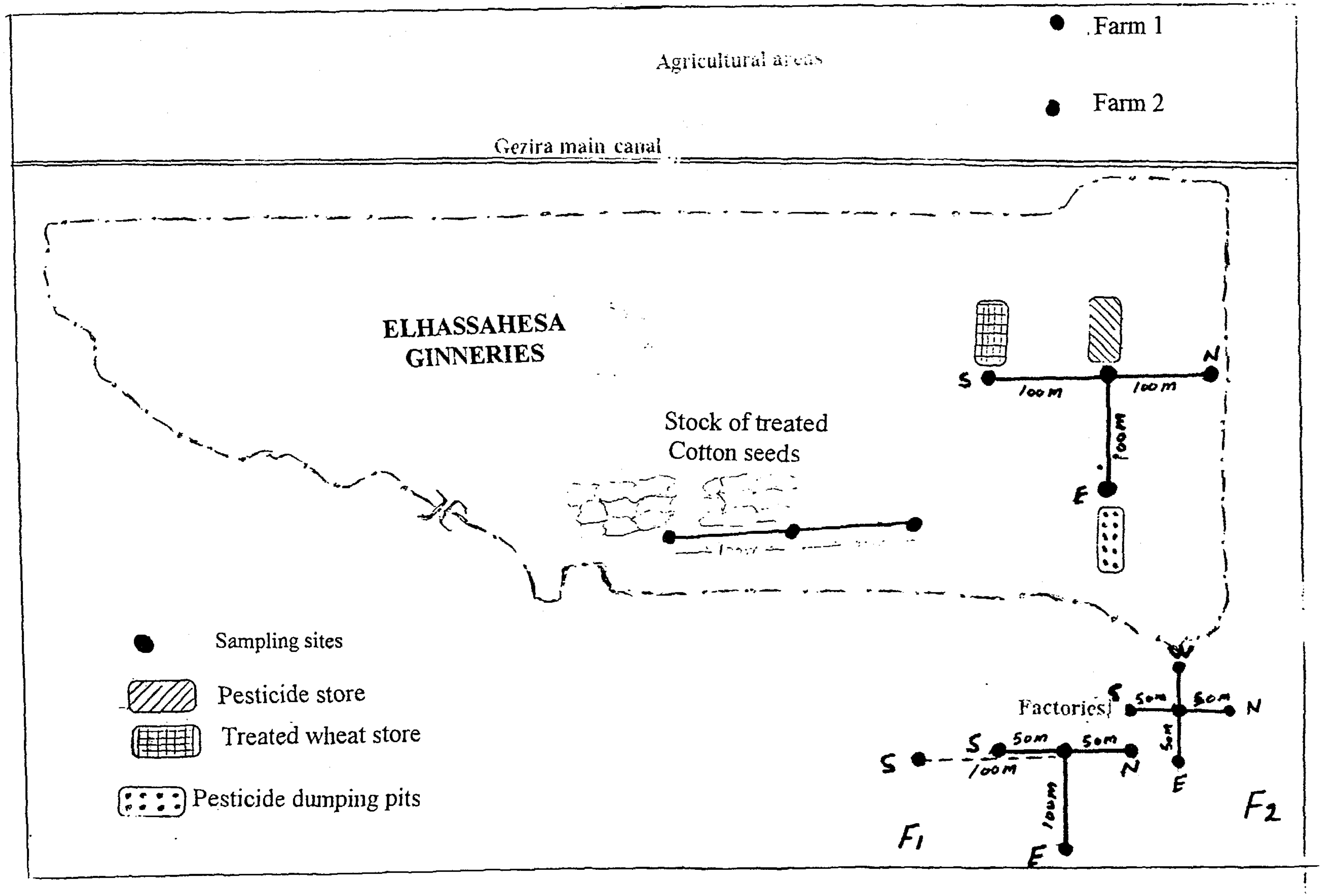

Fig. 5b: Location of sampling sites (Detailed)

ture, University of Khartoum for analysis of soil physical and chemical properties

3.3 X-ray fluorescence (XRF) analysis

The continuing interest in trace elements has stimulated the development of several powerful analytical techniques for their detection and quantitative determination. One of these techniques is the X-ray fluorescence (XRF), where a source of X-ray photons is used to study the elemental composition of materials. The methods gives information about the element present in the sample irrespective of their chemical structure or phases in which they exist.

In this study, the XRF technique was used for determination of total mercury concentration in environmental soil samples collected from Elhassahesa area.

There are three stages for quantitative XRF analysis of a sample viz; preparation of specimen; excitation of a suitable emission line of the element of interest; measurement of their intensity and determination of their concentrations through a suitable calibration. The soil samples were crushed into fine powder. About one gram of the powder from each sample was then pressed into pellet form (2.5 cm diameter) using pressing machine. Pellets were then presented to the XRF spectrometer for measurement of total merucry. The characteristic X-rays (of the elements present in the sample) produced during the irradiation were detected by a Si (Li) detector.

The spectra obtained on a multi-channel analyzer (MCA) were transferred directly to a computer where the X-ray spectrum was analyzed. Sketch of the XRF spectrometer system is shown in Fig (6). The spectra were first analyzed using a program called Analysis of X-ray spectra by iterative least square fitting (AXIL). The AXIL program separates overlapping peaks; identifies the elements and determines the net area of the peaks which are proportional to the concentrations of elements in the samples. A sub-program in AXIL called QAES (quantitative analysis of environmental samples) was used for the quantitative determination of the elements present in the samples.

A standard soil, called soil 7, supplied by the IAEA (International Atomic Energy Agency) of Vienna, was used for calibration of the XRF system and to ensure reliable results.

3.4 Soil chemical and physical properties

One soil sample was chosen from each location for the determination of soil pH, organic matter content, water soluble cation and anion, texture and structure. The soil chemical analysis was made according to Page *et al.* (1982), while the physical analysis was made according to Black *et al.* (1965).

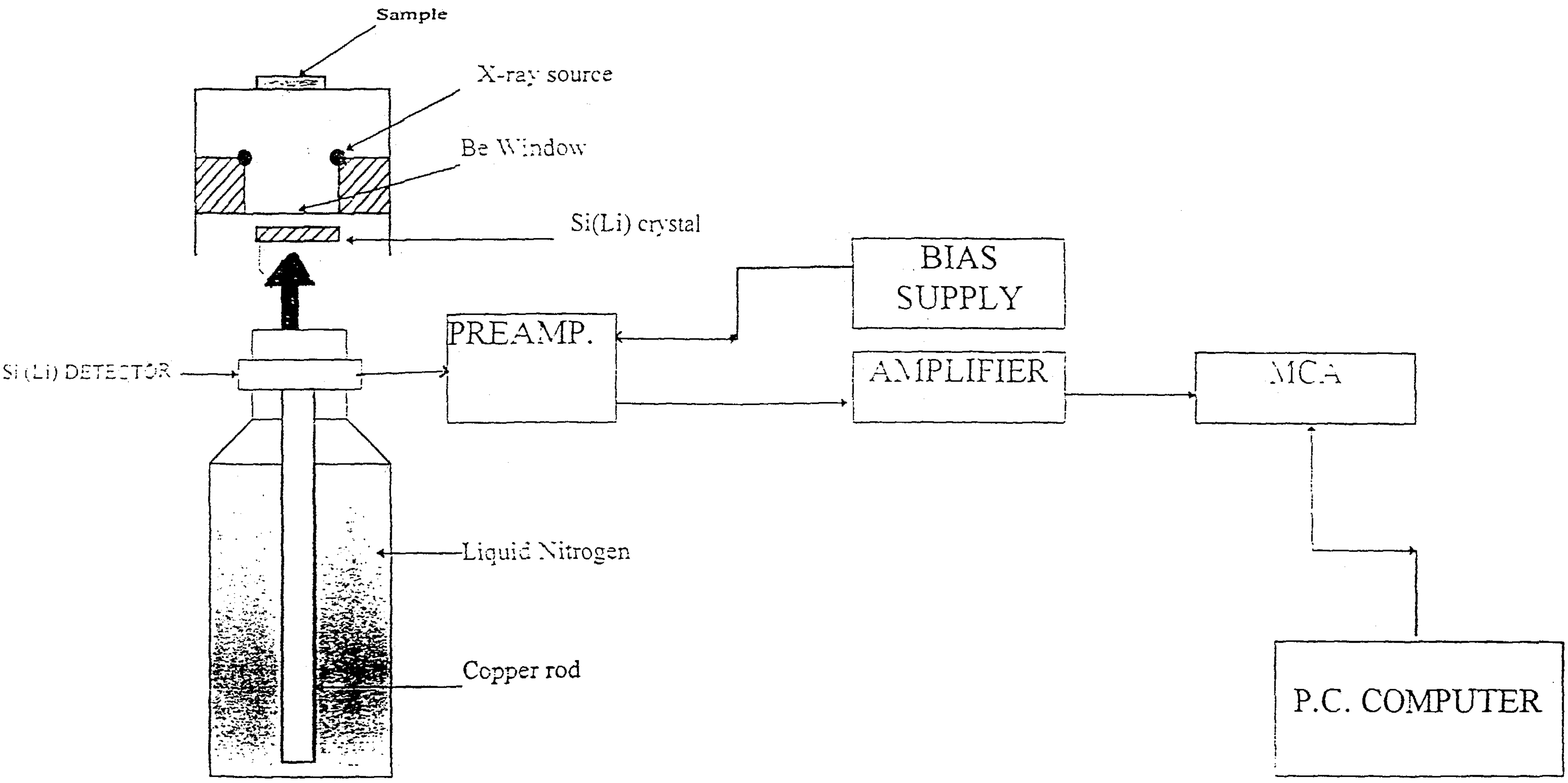

Fig 6: Si(Li) Spectrometer System.

CHAPTER FOUR

RESULTS

Surface soil samples were taken from five locations in Hassahesa town, Gezira State, Sudan. The samples taken represent soils from pesticide store area, food processing factories whcih used treated cotton seed as fuel, near stock of treated cotton seeds and from the neighbouring cotton fields. Soil was sampled at variable distances (0 m, 50 m and 100 m) and directions from the foci of these locations. Control was taken from soil of similar properties from Arqueet areas. Samples were analyzed using XRF spectrometer for determination of total mercury according to the methods of Piorek, 1978). Typical X-ray spectrum is shown in Fig. 7.

Physical and chemical properties of the soil were determined from composite samples of each location according to the methods of Page *et al.* (1982) and Black *et al.* (1965)

The results indicated high levels of contamination with mercurials in the areas studied far greater than the corresponding level of control soil.

The highest level of contamination was detected in samples from pesticide store (average 51 ppm and range 11.5-185 ppm), followed by the two food processing factories (average 24 ppm and range 8.2-72.5 ppm for factory I; 19 ppm and 9.6-36.0 ppm for factory II), soils near stock of treated

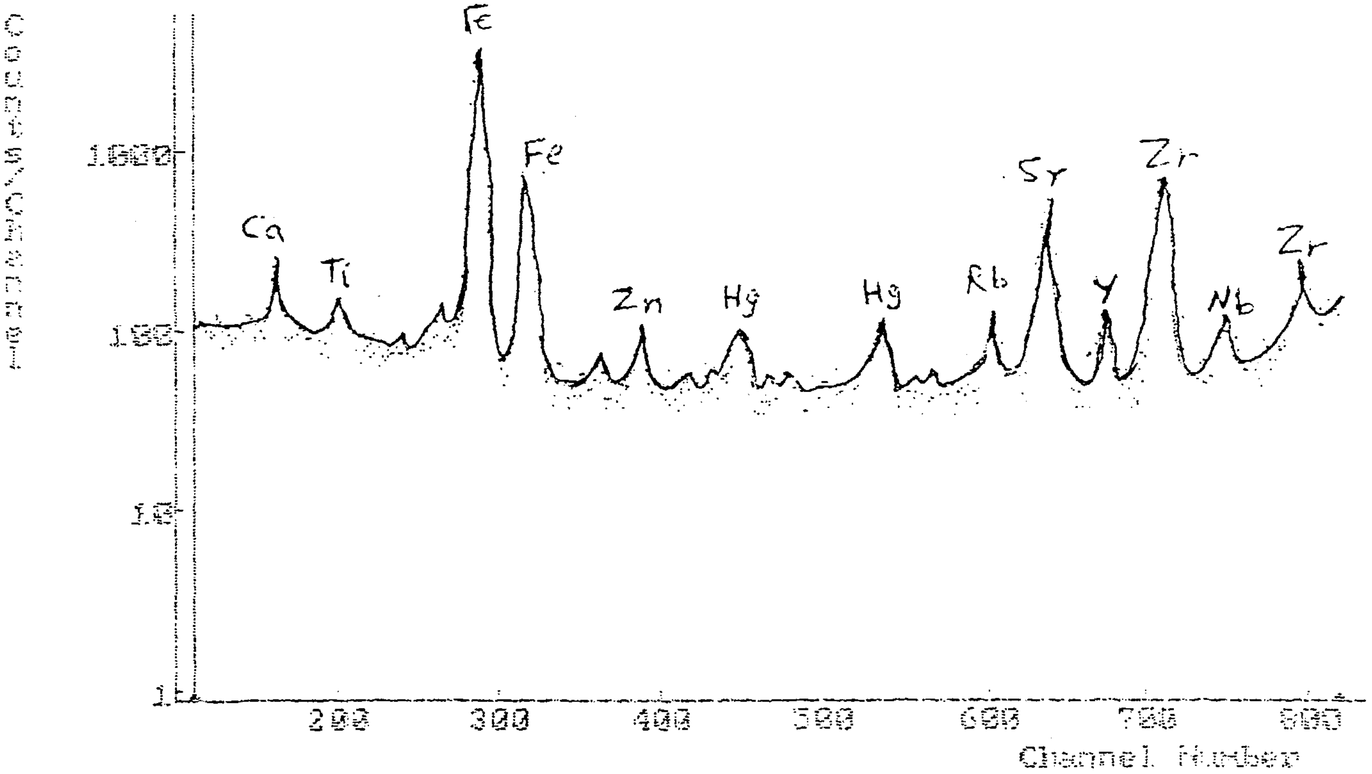

Fig.7: Typical X-Ray spectrum.

cotton seeds (average 15 ppm and range 11.7-21.4 ppm) and lastly soil from neighbouring cotton fields (average 14 ppm and range 11.8-16.4 ppm) [Table 7]. Detailed data on the level of total mercury in the different sites were summarized below:-

4.1 Level of total mercury in the soil of pesticide store

Table 8, Fig. 8 and Appendix I indicated that the highest levels of contamination with mercury was found at the origin of the store where the average concentration of total mercury is 154.67 ppm. This level was the highest compared to the other samples tested. Level of contamination with mercury decreased progressively with distance from the origin of the store and approaches 21.23 ppm at 100 meter south, 16.1 ppm at 100 meter north and 12.77 ppm at 100 meter east. No samples were taken at 100 meter west because this direction was covered by storage-shed.

4.2 Level of total mercury in the soils of food factory I

The results summarized in Table 8, Fig. 9 and Appendix II indicated that the highest level of contamination with mercury was detected at 50 meter south the factory (45.9 ppm), followed by 50 m north (32.6 ppm), origin of the combusion chamber (14.8 ppm), 100 m east (14.63 ppm) and 100 m south (13.4 ppm).

Table 7: Range and average concentration of total mercury (ppm) in soils of the five locations studied

Locations	Samples analyzed	Average	±SD	Range
Pesticide store	12	51.1916	64.2383	11.5-185
Factory I	15	24.2667	16.3185	8.2-72.5
Factory II	15	18.86	8.7508	9.6-36
Stock of treated cotton seeds	9	15.4773	3.8002	11.7-21.4
Cotton fields	8	13.5	1.6212	11.8-16.4
Control soil	3	11.03	-	-

SD: Standard deviation.

Table 8: The average concentration (ppm) of the total mercury in the soil of the five locations studied

Location	Direction	Average conc.±SE
	Origin	154.66±19.5982
Pesticide	East (100 m)	12.766±0.72640
store	North (100 m)	16.1±2.4193
	South (100 m)	21.233±4.32120
	Origin	14.8±2.4006
	East (100 m)	14.633±0.4095
Factory I	North (50 m)	32.6±3.5387
	South (50 m)	45.9±13.3097
	South (100 m)	13.4±2.6005
	Origin	13.966±2.8414
	East (50 m)	15.4±1.3076
Factory II	West (50 m)	15.9±0.9073
	North (50 m)	14.066±1.9741
	South (50 m)	34.966±0.7964
Stock of	Origin	16.833±2.5206
treated cotton	Origin	15.033±2.8948
seeds	North (100 m)	14.566±2.9340
Cotton fields:		
Farm 1		13.25±0.9318
Farm 2		13.75±1.4542
Control		11.03

SE: Standard error.

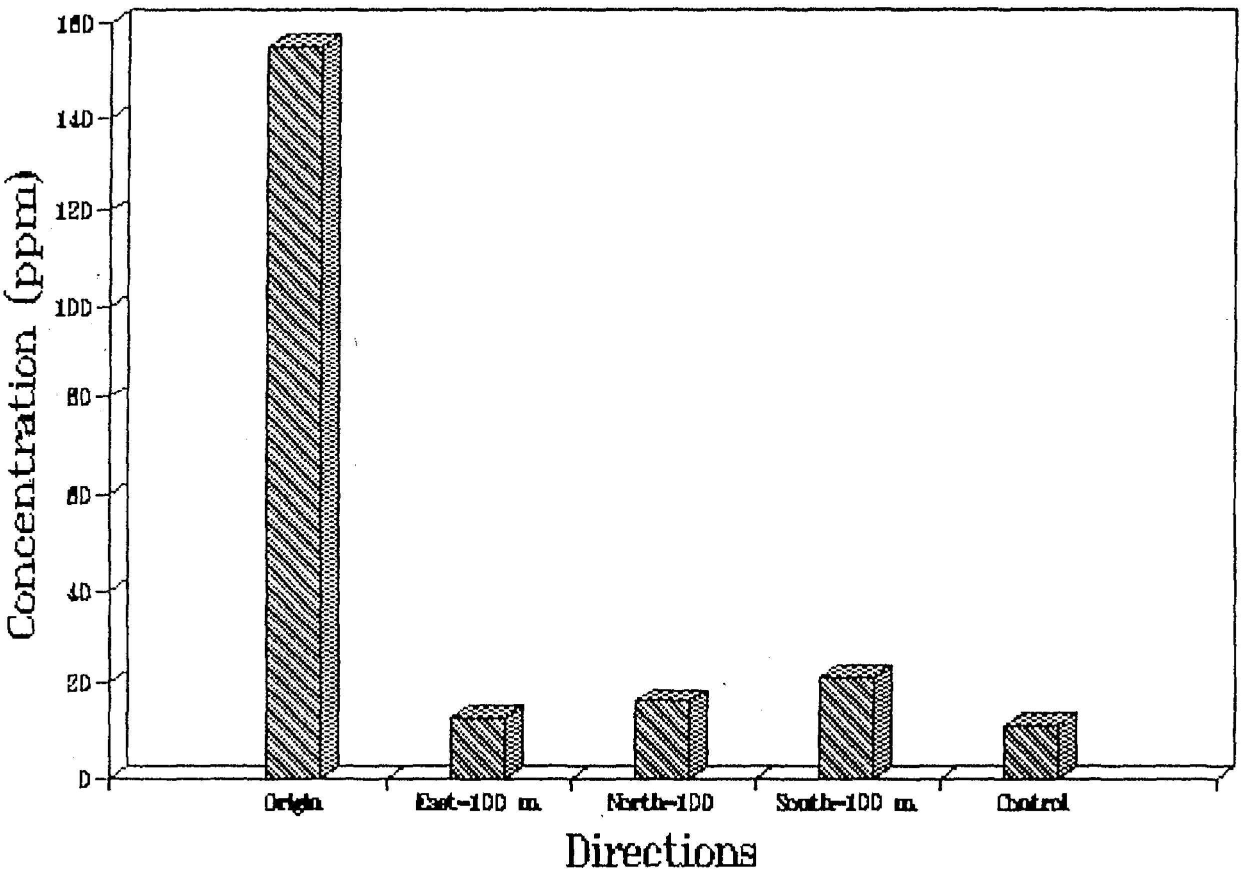

Fig. 8: The concentration (ppm) of total mercury in the soil of the pesticide store

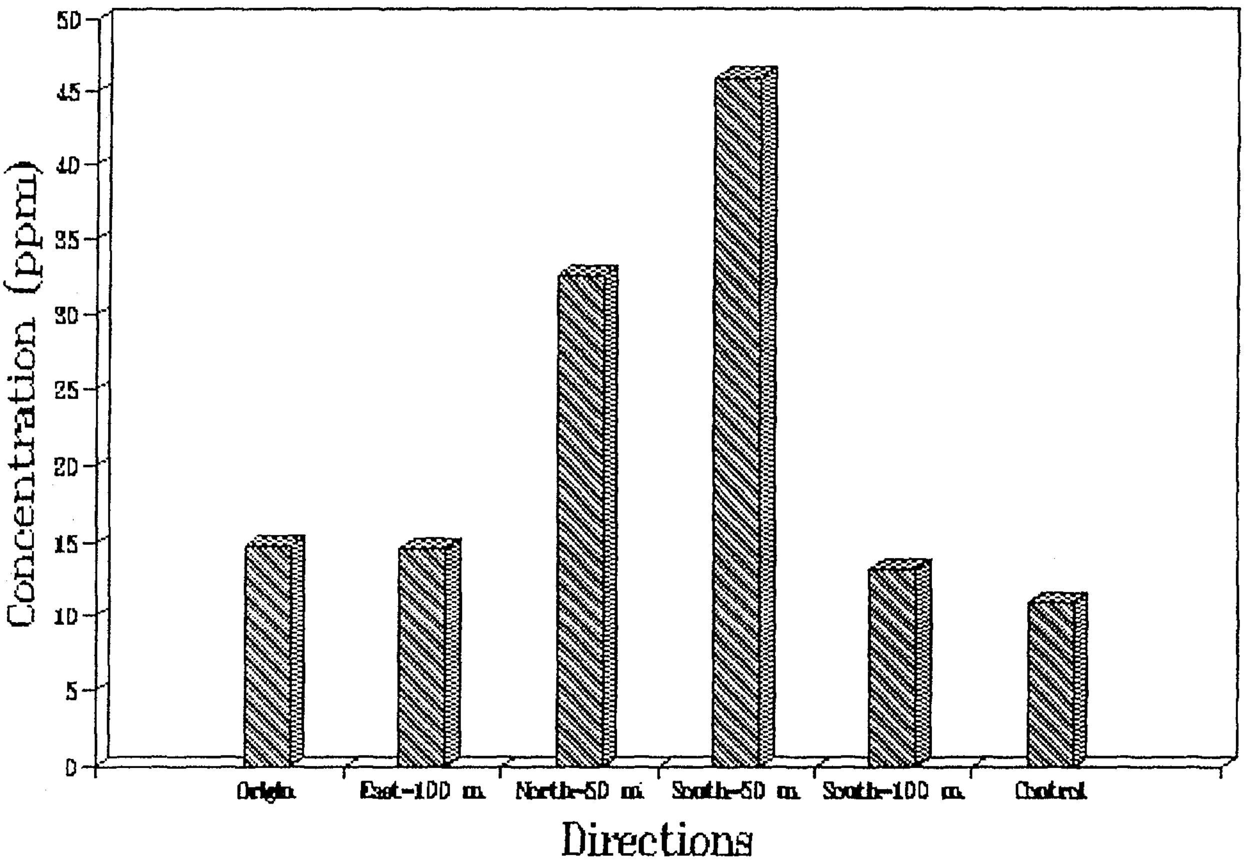

Fig. 9: The concentration (ppm) of total mercury in the soil of factory 1

4.3 Level of total mercury in the soils of food factory II

Results summarized in Table 8, Fig. 10 and Appendix III showed that the highest level of total mercury was detected 50 m south the factory (34.97 ppm), followed by lower values at 50 m west (15.9 ppm), 50 m east (15.4 ppm), 50 m north (14.06 ppm) and the origin of combusion chamber (13.96 ppm).

4.4 Level of total mercury detected in the soils near the stock of treated cotton seeds

Results of total mercury in the soils near the stock of treated cotton seeds were summarized in Table 8, Fig. 11 and Appendix IV. The highest level was detected in soil closer to the border of the seed sacs (16.833 and 15.033 ppm). The total mercury concentration at the third sampling site (100 m north the sacs) contains a relatively lower level (14.57 ppm).

4.5 Level of total mercury in soils of residential area and normal cotton fields

Level of total mercury in soil of two cotton fields was found as 13.75 and 13.25 ppm [Table 8, Fig. 12 and Appendix V].

Corresponding level in soil from residential area south Hassahesa town was found as 11.03 ppm.

Physical and chemical properties of the soil were determined from composite samples of each location (Table 9).

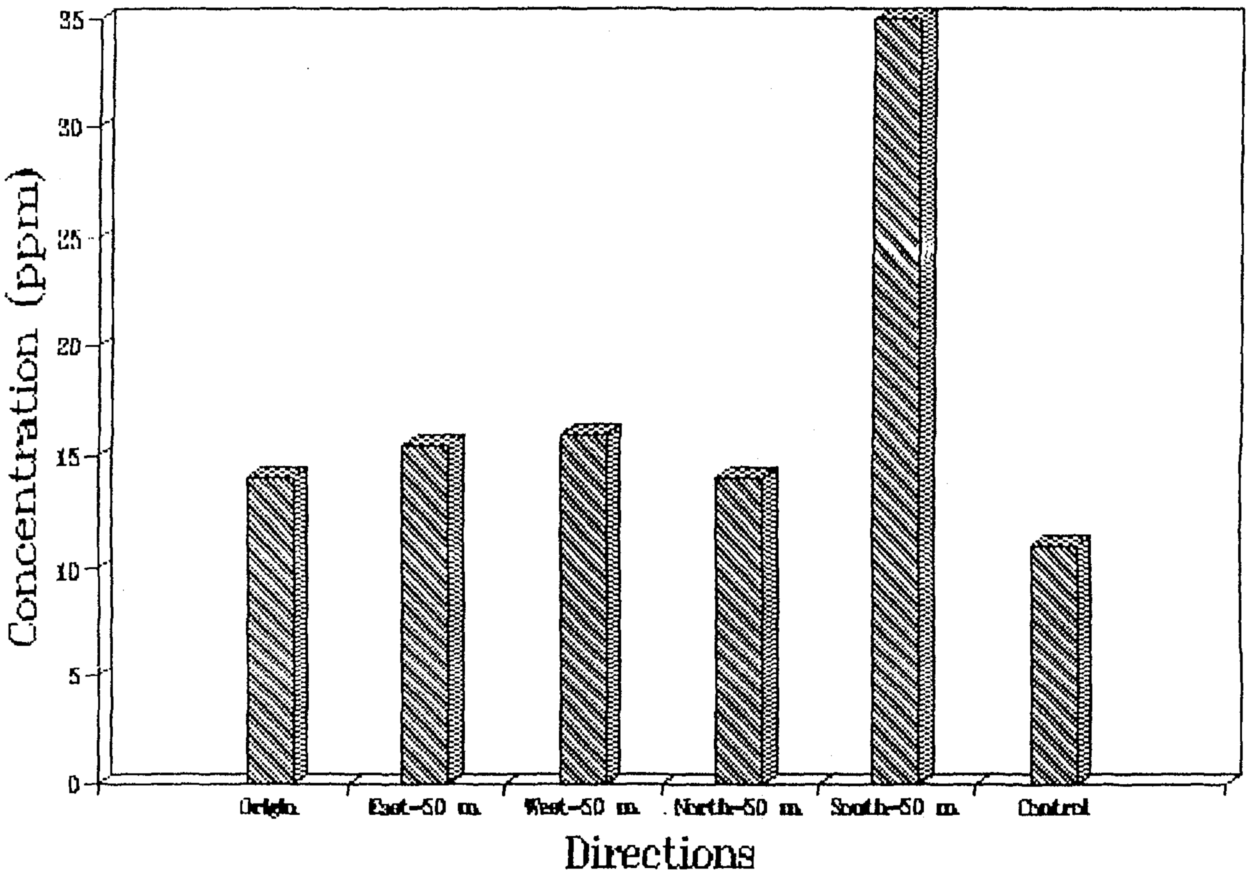

Fig. 10: The concentration (ppm) of total mercury in the soil of factory II

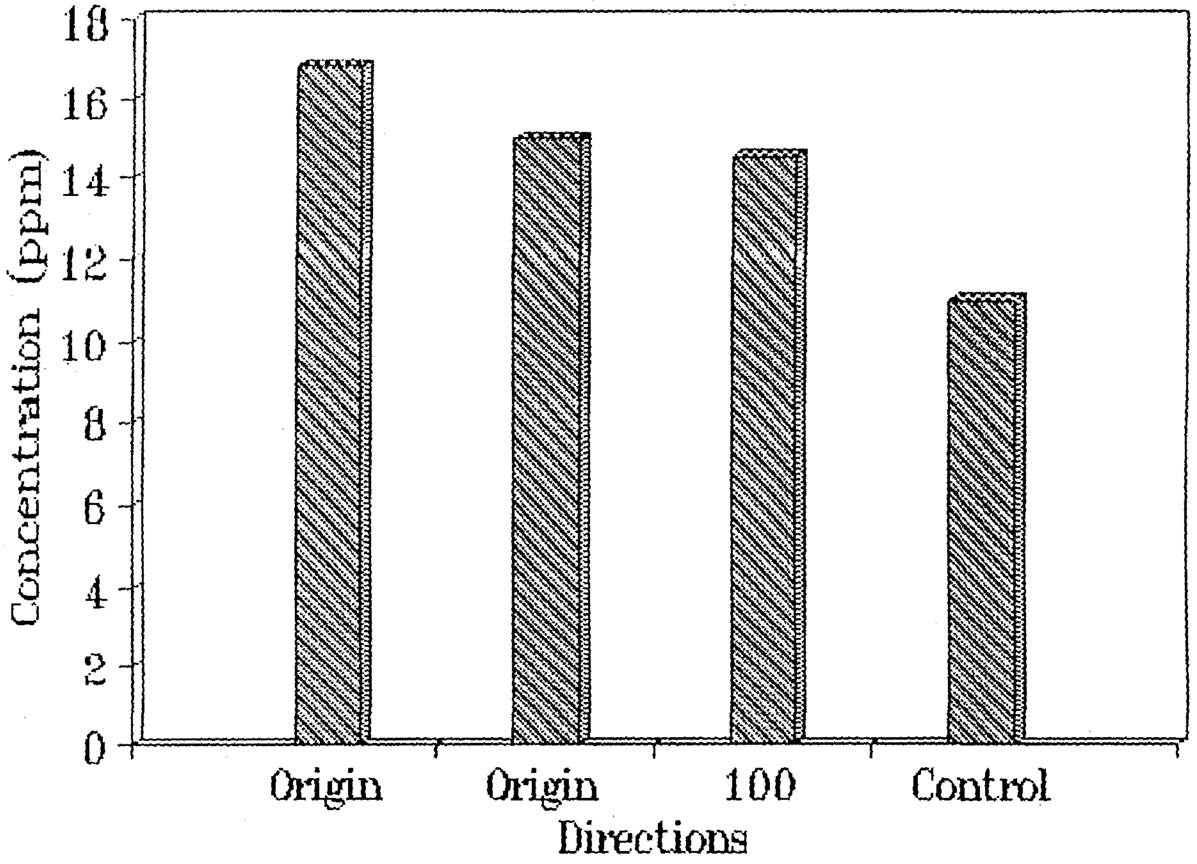

Fig. 11: The concentration (ppm) of total merucry in the soil from near stock of treated cotton seeds

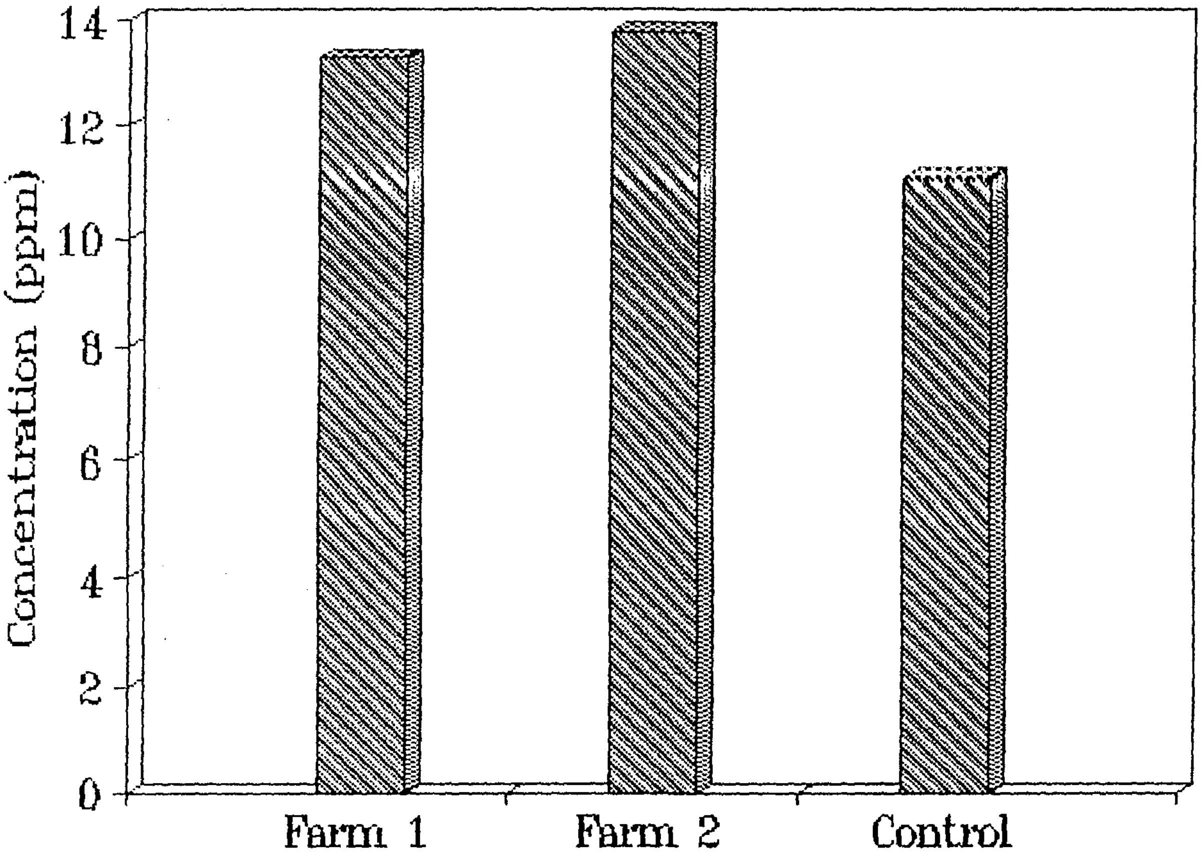

Fig. 12: The concentration (ppm) of total mercury in the soil from near by cotton fields

Table 9: Soil chemical and physical properties

Parameter	Locations			
	Pesticide store	Factories	Near stock of treated cotton seeds	Cotton fields
Electrical conductivity	17.4	11.6	1.32	1.1
pH (paste)	5.5	6.7	7.4	7.2
$Ca^{++} + Mg^{++}$ (meq/L)	290	11	22	18
Ca^{++} (meq/L)	136	107	16	14
Na^{+} (meq/L)	0.34	0.45	0.14	0.12
K^{+} (meq/L)	0.15	0.03	0.01	0.01
$HCO3^{-}$ (meq/L)	9	5	15	12
Organic matter %	1.2	3.1	2.9	3.6
CEC meq/100 g soil	36.8	26	47.5	54.3
Clay %	55.25	22.75	62.75	62.75
Silt %	4.75	12.50	22.55	17.5
Sand %	40	64.75	14.75	19.75
Texture	Clay	Clay loam	Clay	Clay

CEC: Cation exchange capcitiy.

CHAPTER FIVE

DISCUSSION

Mercury fungicides are of historical and classical toxicological interest. Hundreds of compounds of aryl mercurials have been synthesized, but only those containing a phenyl group are of major importance, especially as pesticides and biocides. Phenyl mercury acetate (Agrosan) is the most widely used of the aryl mercurials (Goldwater and Stopford, 1977).

The toxicological hazards associated with the use of organomercuries are well known. Because of their persistence, organomercurial get into the food chain, so several countries have now banned their use (Nene and Thapliyal, 1979) and most registration for mercury fungicides were canceled by EPA. This decision hinged on their toxicity to warm-blooded animals and accumulation of mercury in the environment (Ware, 1983).

In Sudan, seed dressing, especially for cotton seeds, has been strictly adopted since last thirties for the control of black arm disease caused by *Xanthomonas malvacearum*. The first chemical used was abavit [mercuric chloride + mercuric iodide] (Saad, 1975). In 1980, Agrosan/heptachlor was introduced into Gezira Scheme and continue till 1994 where it was replaced by Agrosan/Lindane. The latter continued in practice until 1997 where it was replaced by Bronopol mixed with supersan (season 1997/98). Currently, since 1998 a mixture of three chemicals, Apron 200 LS (1cm^3) + Quinolate 150 plus (2 cm^3) + Promet 400 CS (4 cm^3)/kg seeds is anually used. The annual import of Agrosan during the period of its active use was estimated at 2500 Kg which are enough for treating about 4000 tons of seeds (enough for growing 300,000 feddans) [Seed Propagation Dept., personal communication).

The banning of this chemical in 1994 and latter in 1997 (Sudan) coupled with poor management practices at both storage and application levels resulted in huge piles of inviable treated cotton seeds. These intoxicated seeds were stored in open at the poorly fenced area of cotton ginneries at Hassahesa and Maringan. This situation led the authorities in Gezira scheme to sell significant amount of these intoxicated seeds as fuel to local food factories in Hassahesa town. The accident received great criticism from both local public and environmental authorities.

Mercury vapour and/or oxides may emanate from chimneys of these factories and settle down in the nearby area and therefore pose a serious threat to environmental quality there.

This situation initiated our interest to investigate the levels and movement of mercury compounds in the affected sites. Such study is badly needed for any future environmental assessment study about the accident. The significance of this study also stems from the toxicological and environmental hazards associated with high level of mercurials in the environment.

The results indicated high levels of contamination with mercury compounds in all areas studies (including the control soil), far greater than the corresponding background and normal range of mercury in soil reported by Saha (1972) and Goldwater and Stopford (1977).

The highest levels of contamination was detected in samples from pesticide store followed by the two food processing factories, soil near the stock of treated cotton seeds and soils from the neighbouring cotton fields.

The level of soil contamination is seriously high at the origin of the pesticide store (155 ppm) and decreases progressively as we go away in various directions. It is worth to mention that the origin of the store is almost covered with scattered treated cotton seeds which decreases in abundance as we go in various directions. The level of total mercury 100 meter south of the store showed slight elevation and according to an interview of the store keeper, this site was used previously for storing wheat seeds treated with Agrosan.

Elevated level of total mercury was also found in certain spots in the two food factories at 50 meter south the combusion chambers of both factories (46 ppm and 35 ppm for factory I and II respectively) with relatively lower in the foci of combusion chambers and other direction sampled. The north direction which confined with the axis of the seasonal wind direction (south east) had higher level (33 ppm at 50 meter north, factory I) related to other directions. The interview with the working staff in both factories revealed that the 50 meter south the combustion chamber area was designated for ash dumping after regular cleaning of chambers. This could explain the higher levels observed in these spots.

Mercurial contaminants detected near the stock of treated cotton seeds showed lower levels compared to the pesticide store and the food factories. Samples in north direction could received further contaminants by the action of wind, run off rain water as they are confined within a low land area. The movements of surface soil contaminants by the action of wind, wind blown dust, run off rain water was reported by several authors (Ashton, 1961; Edwards *et al.*, 1970; Lichtenstein, 1970; Cohen and Pinkerton, 1966). Soil surface movements of pesticides are usually accelerated by steep topography; low soil permeability; considerable rainfall and strong adsorption of pesticides to particles on or near the soil surface (Le Grand, 1966; Peach *et al.*, 1973). Therefore, this could provide a further explanation to the relatively greater movement of mercury contaminants to the north direction.

No attempt was done towards evaluation of downwards movement because of time shortage, but it should be investigated in future line of research together with levels in other environmental compartments such as water, air, food crops, animals, human ... etc. The level of mercury in the products of these factories is of equal significance and should be carefully investigated in future studies as well.

Previous laboratory work on combusion of treated cotton seeds (treated with Agrosan) reported the possibility of liberation of toxic gases to the atmosphere such as mercury vapour, Co, So_2 and Tolwin which may endanger the human health and lead to air and soil [by condensed mercury vapour] pollution (Elmaheina, 1995).

Mercury fungicides are toxic to all forms of life. As a result, no mercury residues are permitted in food or feed (Ware, 1983). Its salts are poisonous, particularly the fat-soluble methyl mercurials and water soluble mercury (II) salts. Mercury metal is less toxic because of its small solubility when swallowed, but the vapour is dangerous to inhale.

The inadvertent ingestion of seed treated with organic mercury as a fungicide is a hazard, despite a coloured dye being used and the container marked in several languages (Hutton, 1987). There have been several incidents of mercury pollution with lethal consequences, such as Iraq epidemic in 1971-1972. This epidemic resulted from consumption of bread prepared from grain dressed with alkyl mercury fungicides. The incident in question resulted in the poisoning of about 6000 individual (Clarkson, 1977) and death of over 500 in hospital (Bakir *et al.*, 1973).

Levels of total mercury in cotton fields and control soil were high and far exceeding the background and normal range of mercury in soil (Saha, 1972; Goldwater and Stopford, 1977).

Elevation of total mercury level in cotton fields which were regularly grown from treated cotton seed (with Agrosan) is expected.

No data of the background level of mercury in the Sudanese soil was available. The high level of total mercury in the control soil of this study is very strange and because of the limited number of samples and/or the method of analysis we could not speculate upon at this stage, further studies with greater number of samples from various soil types and more advance method of analysis is needed for clarification of this point. No correlations has been done between soil physical, chemical properties and the residue levels due to the limited number of samples. But there are many soil factors known to influence the behaviour and fate of organomercury fungicides in soil (Mcewen and Stephenson, 1979). The chemical conversion from organo forms to the biologically toxic mercury occurs by a base exchange reaction involving an organomercury-clay complex as an intermediate. A higher clay content might actually result in greater chemical degradation but more pesticidal activity (Booer, 1944).

The reaction of organic and inorganic mercury compounds with soil organic matter leads to volatilization of Hg° state (Hitchcook and Zimmerman, 1957). Landa (1978) suggested that soil microorganisms mediate the volatilization of Hg° following the reduction of various mercury compounds and organomercury complexes to Hg° in soils, sediments and surface waters.

The vapour is lost to the atmosphere, where it apparently remains in this same form (Slemr *et al.*, 1981; Fitzgerald *et al.*, 1982). The fact that central clay plain soil is known for its high clay content may contribute to elevated level of active mercury compounds and this may endanger the human health.

The results clearly indicated an elevated mercury level in all samples analyzed far exceeding the background and normal range of mercury in soil. Several points need further investigations as indicated previously. The possibility of cleaning the affected sites by chemical treatment, by washing with acidic solution and complexation with added resin or by flushing with chelating agent could help in cleaning these such site (Tuin and Tels, 1991; Peters and Shem, 1992) and therefore deserve critical investigations.

CHAPTER SIX

CONCLUSION AND RECOMMENDATIONS

1] Soil from various sites investigated (including cotton fields and control soil) showed an elevated level of total mercury far exceeding the standards level.

2] The highest level of contaminants was found in the soil of the pesticide store followed by the two food factories and the stock of treated cotton seeds.

3] Horizontal movement of mercury contamination was observed and more clear in site confines with the axis of seasonal wind current and/or low land.

4] The danger of mercury dressed grains being eaten by animals rather than planted and the danger of eating these animals or using their milk or products necessiate the need for intensive programme aiming at the decontamination of soil and environment as well as disposal of the existing redundant stocks.

5] An environmental monitoring programs should be launched immediately to reveal the level of mercury contamination in various environmental compartments (soil, water, food, products of factories ... etc) in the area covered.

6] Regulatory laws aiming at no mercury contamination is allowed in food or feed as well minimum pollution to the environment should be strictly applied. In this connection import and usage of pesticides containing heavy metals specially mercury should be restricted or banned. Awareness of public and decision-makers about the toxic effects of pesticides and their hazards to public health and environment should be raised through various types of extension programs.

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Appendix I: The concentration (ppm) of total mercury in the soil of pesticide store

Sample Number	Directions			
	Origin	East (100 m)	North (100 m)	South (100 m)
1	161.0	14.1	19.7	29.4
2	118.0	12.6	17.1	19.6
3	185.0	11.6	11.5	14.7
Mean	154.66	12.766	16.1	21.233
SD±	33.945	1.2583	4.1905	7.4848

S.D: Standrad deviation.

Appendix II: *The concentration (ppm) of total mercury in the soil of factory "I"*

Sample Number	Directions				
	Origin	East 100 m	North 50 m	South 50 m	South 100 m
1	19.6	15.4	39.4	33.5	16.1
2	12.5	14.5	30.9	31.7	15.9
3	12.3	14.0	27.5	72.5	08.2
Mean	14.8	14.633	32.6	45.9	13.4
S.D±	4.1581	0.7093	6.1294	23.0538	4.5044

S.D: Standrad deviation.

Appendix III: The concentration (ppm) of total mercury in the soil of factory "II"

Sample Number	Directions				
	Origin	East 50 m	West 50 m	North 50 m	South 50 m
1	19.3	17.5	17.7	18.0	35.5
2	13.0	15.7	15.2	12.4	33.4
3	09.6	13.0	14.8	11.8	36.0
Mean	13.966	15.400	15.9	14.066	34.966
S.D±	4.9217	2.2649	1.5716	3.4195	1.3795

S.D: Standrad deviation.

Appendix IV: The concentration (ppm) of total mercury in the soil near the stock of treated cotton seeds

Sample	Directions (south ——————> north)		
Number	Origin	Origin	100 m
1	21.4	20.8	18.0
2	16.4	12.6	13.5
3	12.7	11.7	12.2
Mean	16.833	15.033	14.566
S.D±	4.3661	5.01429	3.0435

S.D: Standrad deviation.

Appendix V: **The concentration (ppm) of total mercury in the soil of neighbouring cotton fields and residential area**

Sample Number	Farm 1	Farm 2	Residential area
1	15.2	16.4	11.3
2	12.9	14.3	11.6
3	12.5	12.5	10.2
4	12.4	11.8	10.2
Mean	13.25	13.75	11.03
S.D±	1.6140	2.5189	0.0

S.D: Standrad deviation.