



Assessment of Groundwater Contamination of the Northern Part of the Sana'a Basin along the Sewage Canal with Pesticide Residues.

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ABSTRACT:

The excessive use of pesticides by many farmers, using pesticides that do not belong to public health pesticides in agriculture and Qat production, and the pesticide dealers' violation of the controls followed in the illegal supply of pesticides in many areas of Yemen, led to major violations in the use of pesticides by farmers and the disposal of pesticides. Improper waste of these pesticides eventually led to the contamination of water bodies and groundwater with many types of pesticides. On the other hand, pesticide residues are disposed of in an improper manner, as pesticide residues are poured into the sewage network and seep into the lower ground layers, which contributes to pollution. Groundwater with pesticides. This study aimed to determine the effect of wastewater on groundwater contamination with pesticides in the northern part of the Sana'a Basin, where 42 samples were collected from groundwater wells along the sewage canal in the study area. Samples were prepared for analysis by extracting water samples using N-Hexane solution, and pesticide residues were analyzed using ICP Varian-450 induction plasma (GC-ECD) technology. Four types of pesticides appeared in 5 wells 1, 14, 22, 3 and 33: Tolclovos, Thiamethoxam, and Cypermethrin at concentrations of 0.0040, 0.0014, and 0.0056 ppm, respectively. Lambda-cyhalothrin pesticides also appeared in Well 3 and Well 33 at concentrations of 0.000025 and 0.0039 parts per million, respectively. These results indicate that this water is not fit for use due to the resulting health and environmental damage. A comprehensive study of the studied area must be conducted to determine the main causes of pollution, enact strict laws and regulations to reduce pollution, and monitor the condition of groundwater to ensure that no pesticide contamination occurs, since groundwater is the main source of drinking water.

Introduction

Water is a source of life for life. This substance must be preserved from all influences: modern civilization, industrialization, urbanization, and an increase of the population¹. Groundwater may be contaminated and contain elements that may have adverse health effects, such as pathogenic microorganisms, hazardous chemicals such as pesticides or even toxic elements and compounds³. The groundwater contamination in an urban environment is due to the infiltration of domestic untreated wastewater

into the natural receiving environment, accentuated by favorable hydrogeological conditions².

Pesticides have an extended history of widespread use worldwide. In spite of their proscription and restrictions, many countries are yet using them for various intentions (public health, agriculture), because of the low cost, ease of use, versatility in controlling various insects^{4,38}.

Using of pesticides is ubiquitous in modern agriculture practices. An immense quantity of these chemicals pertains to crops grown by farmers all over



the globe. World consumption of pesticides is increasing, and the situation is getting worst. Across the world, pesticide consumption is close to 2 million tons per year ⁵. A direct movement of pesticides into groundwater is a common type of point source pollution, in which the pesticides enter the water wells result from pesticide spills and improper disposal of pesticides ⁶.

Pesticides are widely sprayed in gardens, agricultural lands, and other public areas to kill undesirable organisms. Also, expansion in agricultural activities leads to excessive use of different pesticides. These pesticides present on crops and in fields get washed with rainwater from agricultural areas and get collected in water bodies ⁷. Pesticides pass through the soil and subsoil, resulting in groundwater contamination, in addition to agricultural activities, wastewater produced during vegetable washing in vegetable processing facilities and pesticide manufacturing units contains pesticides way beyond permissible limits. This wastewater reaches water bodies through drains and by other means and result in groundwater contamination ⁸⁻⁹⁻¹⁰.

It is recognized that effluent wastewater is one of the main sources of groundwater pollution and usually contains high concentrations of micropollutants ¹¹⁻¹². Agricultural runoff, industrial discharges, sewage systems, and crop irrigation with reclaimed wastewater ¹³. The most important factors causing pesticide contamination of groundwater ¹⁴.

Factors influencing pesticide movement to groundwater include properties of the pesticide, properties of the soil, rainfall and depth of ground water, method and rate of application of pesticide ¹⁵. Many of these pesticides and their metabolites have been involved in a wide range of toxic and adverse human and environmental effects including reproduction and birth defects ¹⁶⁻¹⁷, immune system malfunction ¹⁸, endocrine disruptions ¹⁹, testicular tumors and lower sperm counts in humans ²⁰⁻²¹, human breast and liver cancers ²², altering the metabolism of xenobiotics and steroid hormones ²³. Parkinsons diseases ²⁴.

Information about pesticides in drinking water sources from the city of Sana'a is limited, as a previous study was conducted that described the case of groundwater contamination with pesticides in the Sana'a region. This study describe the contamination by pesticides result in groundwater in Sana'a City. The objective of this study was to evaluate the spectrum of pesticides residues in groundwater samples of Sana'a City by analysis with GC-ECD.

Materials and Methods

Study area.

The study was conducted in the northern area of Sana'a Basin located along the sewage channel of the wastewater treatment plant north of the capital Sana'a and on an estimated length of about 20 km north ²⁶. About 95 % of irrigation in the study area depends upon the wastewater produced from the Sana'a Wastewater Treatment Plant (WWTP), with 5 % coming from groundwater (boreholes) that farmers use to directly irrigate the cultivated areas. The reason for this relates to the high cost of well water and the resultant decrease in economic income in comparison to irrigation with sewer water ²⁶⁻²⁷. Wastewater passage in the Sana'a basin starts at the outlet of the Sana'a City's Wastewater Treatment Plant at the northern edge of the Sana'a basin (Arhab and Bani Al-Harith areas). Treated and untreated wastewater flow together in the channel. This channel is about 2.5 m in width and 20 km the length. The passage runs across agricultural land and very critical geological features, including major faults, and encounters significant geological variations, including volcanic and limestone outcrops ²⁸.

Sampling sites.

Sampling wells were selected to cover almost the whole area of the wastewater channel stream in the study area, the sampling sites between latitudes and longitudes that were 29.3656015°N -42.8765015°E and 04413.87410°E - 04413.87550°E, the total number of the wells were 42 governmental and private wells, figure (1).

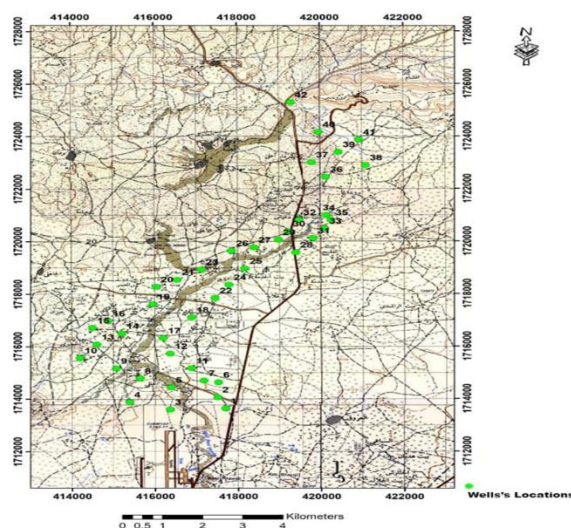


Figure (1): Topographic map of the study area and samples collection sites ²⁹.



To check which pesticides might contaminate groundwater wells, field visits were conducted and farmers were interviewed and asked about the agricultural pesticides they had frequently used in the past years until now. Traders who sell agricultural pesticides were also asked about most of the pesticides sold to farmers, that were frequently and continuously purchased by farmers, and the total number has reached 91 types of commonly used pesticides included: Penconazol, Primiphose-methyl, Propamocarb, Pyridaben, Profenofos, Sulfur, Oxamyl, Teflubenzuron, Temiphos, Tetradifon, Thiacloprid, Thiamethoxam, Thiocyclam, Thiophanate-methyl, Triadimenol, Triadimefon, Triazole, Trifluralin, Tridemorph, Trifloxystrobin, Tolclofos, Flusilazole, Fosetyl aluminum, Fenitrothion, Fenvalerate, Fipronil, Fenpropathrin, Glyphosate, Hexaconazol, Hexathiazox, Imidachloprid, Indoxacarb, Iprodione, Isofenphos-methyl, Kresoxim-methyl, Lambda Cyhalothrin, Lufenoron, Mancozeb, Methrin, Malathion, Metalaxyl, Metiram, Monocrotophose, Myclobutanil, Methiathion, Abamectin, Abtetemephos, Acephate, Aldrin, Azinophos-ethyl, Azinophos-methyl, Azoxystrobin, Acetamipride, Cyfluthrin, Cymoxanil, Cyphenthrin, Chlorpyrifos, Carbendazim, Chlorantraniliprole, Chlorfenapyr, Chlorpyrifos-methyl, Copper Oxychloride, Cycloxydime, Captan, Carbaryl, Chlorothionil, Cyromazine, Cyflumetofen, Alpha-Cypermethrin, Cypermethrin, Deltamethrin, Diaznon, Dichlorvos, Dieldrin, Difenconazol, Diflubenzuron, Diniconazol, Diafenthiuron, Danitol, Dazomet, Dimethoate, Ememectin benzoate, Esfenvalerate, Benomyl, Bifenthrin, Bormocarb, Boscalid, Bifenazate and Bromopropylate.

These pesticides were identified and the pesticides that were unanimously mentioned by farmers and pesticide dealers were approved. There were 48 types of pesticides that were studied, and these pesticides are as follows: Penconazol, Pyridaben, Profenofos, Teflubenzuron, Tetradifon, Thiamethoxam, Thiocyclam, Triazole, Trifloxystrobin, Tolclofo, Fenvalerate, Fipronil, Fenpropathrin, Hexaconazol, Hexathiazox, Imidachloprid, Indoxacarb, Kresoxim-methyl, Lambda Cyhalothrin, Mancozeb, Malathion, Myclobutanil, Methiathion, Abamectin, Aldrin, Azoxystrobin, Acetamipride, Cyfluthrin, Chlorpyrifos, Carbendazim, Chlorfenapyr, Chlorpyrifos-methyl, Copper-Oxychloride, Chlorothionil, Alpha-Cypermethrin, Cypermethrin, Deltamethrin, Diaznon, Dieldrin, Difenconazol, Diniconazol, Dazomet,

Dimethoate, Ememectin benzoate, Bifenthrin, Bormocarb, Boscalid and Bromopropylate.

Analysis of pesticide residue carried out on a Varian- 450 (GC-ECD). Method prescribed by ^{30,33} and with some modifications was used for the extraction of pesticides residue from the water samples. A liquid liquid extraction (LLE) method, using n-hexane as solvent, was used for extraction of pesticide residues. The water samples were prefiltered using 0.45 µm filter to remove any suspended impurity and were extracted without any pH adjustment. For extraction, one liter water sample was 20 taken into a separating flask. It was mixed with 30 g of NaCl and 50ml of n-hexane³¹.

Sample was shaken properly and hexane layer was separated. This process was repeated thrice with same water sample and hexane portions were pooled and transferred to a Buchi Condenser flask through a column of further reduced to 1ml under gentle N₂ stream. The concentrated extract was transferred to airtight GC vials and stored at 20 °C until analysis³¹.

Parameter Conditions: Multy standards was 0.05 ppm, Detector was ECD, Column was Varian Cabillary Column CP-Sil 8 CB (25 m – 0.32mm and film thickness 0.52µm), Temperature of injector and Detector were 250 and 300 °C respectively, Column oven were (90- 250- 280 °C Aat time 0.00- 17.33-32.33 respectively), Column flow was 1.0 mL/min and Carrier and Auxilliary gas was N₂ (99.999%).

The data were analyzed using SPSS (Pearson Correlation) program to study the effect of the depth of the wells and the distance between the studied wells and the source of pollution represented by the sewage channel.

Results and Discussion

Pesticides were analyzed for 42 wells of groundwater samples using GC, and the results were as showed in the tables (1,2) that showed four types of pesticides contaminated of five wells, which were well 1, well 14, well 22, well 3 and well 33. Tolclofos appeared in Well 1 at a concentration of 0.004000 ppm, Thiamethioxam in Well 22 at a concentration of 0.001400 ppm, Cypermthrin in well 14 at a concentration of 0.005600 ppm, and Lambda-Cyhalothrin in well 3 and Well 33 at a concentration of 0.000025 ppm and 0.003900 ppm, respectively, tables (1 and 2).

**Table (1): The concentration and types of the detected pesticides in the studied water samples.**

| Conc. ppm | Area % | Area (µV.Min) | Height (µV) | Quantity (% Area) | Time (Min) | Pesticide & well NO. | Index |
|-----------|---------|---------------|-------------|-------------------|------------|------------------------|-------|
| 0.004 | 60.497 | 1607.1 | 475621.1 | 60.5 | 7.48 | Tolclofos w1 | 1 |
| 0.0014 | 1.899 | 2351.5 | 18111.6 | 1.9 | 8.49 | Thiamethioxam w22 | 2 |
| 0.0056 | 2.71062 | 38472.7 | 172157.5 | 31.06 | 22.32 | Cypermthrin w14 | 3 |
| 0.000025 | 5.222 | 1016 | 162506.2 | 5.22 | 18.29 | Lambda-Cyhalothrin w3 | 4 |
| 0.0039 | 12.255 | 1551.5 | 140110.1 | 12.25 | 18.31 | Lambda-Cyhalothrin w33 | 5 |

Table (2): The concentration and types of the detected pesticides the well samples with their depth and distance (m).

| Well No. & type of Pesticide | Conc. ppm | Depth (m) | Distance (m) |
|------------------------------|-----------|-----------|--------------|
| Tolclofos w1 | 0.004 | 140 | 800 |
| Thiamethioxam w22 | 0.0014 | 50 | 1100 |
| Cypermthrin w14 | 0.0056 | 70 | 600 |
| Lambda-Cyhalothrin w3 | 0.000025 | 90 | 700 |
| Lambda-Cyhalothrin w33 | 0.0039 | 60 | 700 |

The results of this study were similar in terms of the appearance of pesticide residues in groundwater with the study that was carried out by ³² and it was agreed with the study that carried out by ^{33,37}, which

showed the presence of pesticide residues in the groundwater, and thiamethoxam at 0.5 µg of concentration among those pesticides that appeared during their study.

Factors influencing pesticide movement to groundwater include properties of the pesticide, properties of the soil, rainfall and depth of ground water, method and rate of application of pesticide¹⁶. There were several reasons for organochlorine pesticides contamination of groundwater: due to their molecular features³⁶, persistent nature and translocation from agricultural land³⁵ the abundant application and or long half-life period in a neutral environment³⁵, faults in the well casings allowing surface water into the wells, also there could be large fractures in the bedrock which result in relatively fast flow of groundwater and recharge from the surface³⁶.

This study also showed that there is no correlation between pesticide contamination and the depth of the studied wells or the distance between them and the sewage canal, not even between the pesticides that appeared in this study, table (3).

Table (3): The correlation between depth, distance and pesticides of the studied wells.

| Correlations | | Tolclofos | Lambda-Cyhalothrin | Cypermethrin | Thiamethoxam | Depth | Distance |
|--------------------|---------------------|-----------|--------------------|--------------|--------------|-------|----------|
| Tolclofos | Pearson Correlation | 1 | | | | | |
| | Sig. (2-tailed) | | | | | | |
| Lambda-Cyhalothrin | Pearson Correlation | -.024 | 1 | | | | |



| | | | | | | | |
|---------------|---------------------|--------|--------|--------|--------|--------|---|
| | Sig. (2-tailed) | 0.878 | | | | | |
| Cypermethrin | Pearson Correlation | -.024- | -.024- | 1 | | | |
| | Sig. (2-tailed) | 0.878 | 0.878 | | | | |
| Tthiamethoxam | Pearson Correlation | -.024- | -.024- | -.024- | 1 | | |
| | Sig. (2-tailed) | 0.878 | 0.878 | 0.878 | | | |
| Depth | Pearson Correlation | -.042- | -.164- | -.213- | -.263- | 1 | |
| | Sig. (2-tailed) | 0.794 | 0.299 | 0.175 | 0.093 | | |
| Distance | Pearson Correlation | -.090- | -.167- | -.244- | 0.14 | -.134- | 1 |
| | Sig. (2-tailed) | 0.57 | 0.29 | 0.12 | 0.377 | 0.398 | |

The presence of pesticides in the groundwater is a serious indicator of the occurrence of pesticide contamination resulting from contamination with the wastewater that contains pesticides, as well as the use of fertilizer produced by the wastewater treatment plant, which is often saturated with a lot of the pesticides that used to kill the flies during the drying of these fertilizers as their effect remains in the soil, and their accumulation in them. The excessive use of pesticides by farmers plays a large role in contamination of groundwater. Cypermethrin is a pyrethroid pesticide and exposure to this group has been associated with a number of adverse effects, including oxidative stress, inflammation, and DNA damage. These pathophysiological mechanisms may be associated with the development of acute lung injury³⁸. As a result of human metabolism or environmental degradation, thiamethoxam can cross the blood-brain barrier, resulting in effects on humans³⁹. Lambda-cyhalothrin enters the human body or blood through the digestive system, through various foods (plant or animal). It is noted that the bioaccumulation of pesticides in various organs of the human body is the reason behind the toxicity in the tissues of the liver, brain and reproductive organs, which was discovered through various laboratory studies⁴⁰. Tolclophos -methyl has shown no evidence of immunotoxicity. On the other hand, Tolclophos-methyl is likely to have endocrine disrupting properties⁴¹.

Conclusion

This study showed that 5 of the total number of wells studied were contaminated with four types of pesticides: tolclovos, thiamethoxam, cypermethrin, and lambda-cyhalothrin. These wells are considered unfit for use and must be subjected to a number of processes to treat them so that they do not pose a threat to human health.

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Authors' Declaration

- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- No animal studies are present in the manuscript.
- Ethical clearance: The project was approved by the local ethical committee at Sanaa University, Yemen.

Authors' Contribution Statement

S. Al-Hakami collected the samples and conducted the analyses; S. Dawman prepared the scientific paper with H. Aldoais and N. Alshwafi

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