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Research Article
Cyanide Pollution in Different Water Sources in Assiut, Egypt: Levels, Distributions and Health Risk Assessment

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Abstract
Background and Objective: Nowhere has that situation been more complex and challenging than in Egypt, where combinations of water pollution and scarcity are the most important factors impacting on water safety in Egypt. Water pollution by cyanides is an issue threatening water safety and pose a huge risk to both human and animal health. This study aimed to determine the possibility of the existence of cyanide as a critical chemical contaminant in different water sources at Assiut, Egypt. **Materials and Methods:** Concentrations of free cyanidein98 water samples collected from the surface, drinking, irrigation and waste waters have been evaluated by Lovibond Multi direct Photometer. **Results:** The results showed that cyanide level in wastewater (0.2195 ± 0.027 mg L^{-1}) was significantly higher than surface water (0.037 ± 0.017 mg L^{-1}), irrigation water (0.016 ± 0.009 mg L^{-1}) and drinking water (0.016 ± 0.004 mg L^{-1}) ($p<0.05$). Moreover, cyanide levels in wastewater were found to be above the maximum permissible limit imposed by USEPA and WHO. The mean pH values of the different tested water samples were 7.6738 ± 0.05007 in drinking water 7.7543 ± 0.15602 in irrigation water, 7.6214 ± 0.05570 in surface water and 4.4450 ± 0.04500 in wastewater samples. Statistically, in positive water samples for cyanide, a robust significant negative correlation between pH value and free cyanide values were recorded. **Conclusion:** The presence of cyanide in 54.08% of the collected water samples and a higher concentration in waste waters exceeding the permissible limits is a real threat to the health of all living organisms, especially humans and animals. Therefore, the regular estimation of cyanide in different water sources and in biological materials that allow the assessment of risks resulting from environmental exposure to such hazard pollutant is highly recommended.

Key words: Free cyanide, drinking water, wastewater, surface water, irrigation water, chemical contaminants

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Water is essential for the survival of all living organisms and critical to many sectors of the economy. Therefore, contamination of water resources should be a matter of concern to the human and animal health[1]. Globally, chemical pollution of water is a significant concern for the environment and health. Some anions found in water are essential for human and animal health, however many of them are harmful[2].

Among various anions, cyanide is one of the most hazardous and highly toxic chemicals to human beings and animals. Cyanide binds to the vital iron-containing enzymes, i.e., cytochrome oxidase, which is required for cells to respire aerobically[3]. Disruption of cytochrome oxidases resulting in inhibition of cellular respiration, destruct the mitochondrial electron-transport chain and mortality[4,5]. Assimilation of cyanide can result in either acute poisoning, including death or chronic poisoning to human beings and animals. Chronic exposure affects the respiratory, cardiovascular and central nervous system in human and animal bodies[6].

In the aquatic life, freshwater fishes are the most cyanide-sensitive group of aquatic organisms. Free cyanide at a concentration more than 5 μg L^{-1} can cause a negative impact on the swimming and reproduction of freshwater fish, while at a level higher than 20 μg L^{-1}, cyanide induces high mortalities[7,8].

Cyanide enters into water stream from various resources. Anthropogenic activities like gold mining, manufacturing of pharmaceuticals, plastic burning, discharge of steel industry effluent and leachates from hazardous waste dumping sites may elevate the concentration of cyanide in waters[9]. Cyanide can also occur naturally as a metabolite of several microorganisms or in the form of cyanogenic glycosides in many foods items like cassava, almond and sorghum[10].

Cyanide in water streams can find in various forms such as metal-complex cyanide which may be either weak acid dissociable (WAD) or strong-acid dissociable (SAD) cyanide, cyanide salts and free cyanide[11-13]. The WAD refers to cyanide complexes with metals such as cadmium, copper, nickel and zinc, they are readily dissolved under acidic conditions[14,15]. The SAD refers to cyanide-complexes with metals such as cobalt, gold, iron and silver[14,15]. Cyanide salts as sodium, potassium and calcium are quite toxic. It's toxic and lethal oral doses of cyanide compounds generally range from 50-200 mg CN (0.7-2.9 mg kg^{-1}), according to the EPA. Free cyanide refers to the most toxic forms of cyanide, which includes the cyanide anion itself and hydrogen cyanide (HCN) that is a chemical warfare weapon[16].

Owing to its high toxicity to human and animal health[17], many international guidelines regulated the allowable concentration of free cyanide in drinking water. The United States Environmental Protection Agency (USEPA) has set a maximum permissible limit of 200 μg L^{-1} [18], whereas authorities like the World Health Organization (WHO)[19] and Bureau of Indian Standards (BIS)[20] have imposed a lower limit of 50 μg L^{-1}. European Union (EU) has allowed maximum concentration of 70 μg L^{-1} for mineral waters[21]. However, environmental standards are more stringent for the conservation of the ecosystem. Australia and New Zealand, Environmental Conservation Council has set a triggered value of 4 and 2 μg L^{-1} for freshwater and marine water, respectively[22].

At an environmental level, wastewater is one of the most significant environmental problems in Egypt. Some of the wastewater treatment plants receive vast amounts of sewage that exceeding their capacities, resulting in the expulsion of large quantities of untreated wastewater to the nearest drainage or water body. Arab-Almadabegh area, Assiut, Egypt is suffering from wastewater contaminating its entire environment, as it is harbouring the most magnificent wastewater treatment plants at Assiut, these plants are facing many difficulties in dealing with the daily flow of massive amounts of wastewater. There are variable environmental, local, governmental and even media reports discussing this annoying ecological problem. Moreover, many studies concerning water quality parameters, heavy metals concentrations, bacterial and fungal loads of different water sources have been done before, to estimate water quality of this region. However, this study was the first study conducted to determine the levels of free cyanide at different water sources in Arab-Almadabegh area and some other water bodies where untreated wastewater is disposed of.

MATERIALS AND METHODS

Study area: The study was conducted on the region of "Arab-Almadabegh, Assiut Governorate, Egypt. It is a village, located on the ring road at the west of Assiut city, rich in agricultural lands, produce large quantities of vegetables, fruits and various crops that distributed to many markets at the adjacent areas.

According to many previous environmental studies, this region is suffering from a water pollution problem many years ago[23-25]. The origin of the water contamination problem may attribute to the presence of wastewater treatment plants (WWTP) inside the village itself among the agricultural lands, farmer's houses and small animal farms.

Fig. 1: Google earth maps of the area of the study

WWTP receives extra vast amounts of sewage exceeding its capacity of treatment and to limit its inputs of wastes and to receive wastewater amount that is a bit compatible with its capacity the following was observed:

- Directly and in front of the WWTP1, considerable large amounts of untreated wastewater are continuously expulsed into multiple small open canals that flow between the agricultural land. Due to the lack of another clean water source, farmers are using that untreated wastewater as irrigation water for their agricultural land
- One of the essential sewage lines destined for the treatment plant was broken, so as not to reach the WWTP (already suffering from excess amounts of wastewater) and let that large tube continuously emptying a vast quantity of wastewater in Al-Zennar canal. Thousands of acres are irrigated by water from Al-Zennar canal which extends for few kilometers, passing a long distance, till reach the Nile River, where it flows directly (Fig. 1)

The Nile River at that area is currently facing tremendous pressure due to encroachments, discharge of untreated domestic and industrial waste, dumping of solid waste, in addition to the presence of nearby fertilizer and cement factories on the Nile bank.

Sampling: Sampling was done in a way that allows for cyanide traceability in different water sources in the area of study during the period of 4 months from September-December, 2017. Water samples collected from all water sources according to the standard methods for the examination of water and wastewater[26].

Ninety eight water samples were collected and examined for their possible contamination with cyanide. Twenty eight sample from the drinking water, 28 from the surface water, 20 from the irrigation water and 22 from the wastewater.

Every water source was sampled three times. Samples from each water source were collected from multiple sites, every site was represented by numerous sampling points, from each point, an appropriate number of samples were taken according to the size of the water body and the area surrounding it. All water samples were collected under the same measures and in the same manner at each time of sampling.

Drinking water samples included both tap and groundwater were collected from some taps and hand pump wells that were found at villager's houses (sites 1, 2 and 3). Irrigation water samples were collected from some of the small water canals used for irrigation of the agricultural land (sites 4, 5). Surface water samples included samples from both Al-Zennar canal and the Nile River. Al-Zennar canal water samples were collected from different sites, including many points distributed along the canal from the discharging position of the wastewater tubes until its flow into the Nile (sites 6, 7, 8).

Nile River water samples were represented as (site 9) which included 10 pooled samples, collected from various sampling points, serving the surrounding areas of the point of junction with Al-Zennar canal, north, south to it by hundreds of meters and at different distances from the Nile bank in a total area of nearly about 500 m. A boat was used to reach the appropriate far places for a representative water sampling. Wastewater samples (sites 10, 11) were collected from the wastewater treatment plants as follows:

- **WWTP2:** It is located in a relatively distant place from populated places, it works efficiently, with no discharge of the untreated wastewater before complete treatment. Samples were collected from the inside of the WWTP2 after the entire treatment process and from that water which will be directly discharged via the tubular system to the Nile as treated wastewater (site 11)
- **WWTP1:** These samples were collected from the inside of the plant itself after complete treatment of water and from the untreated, uncovered output of WWTP1 that directly situated in front of the WWTP1(sites10)

Water hydrogen ions concentrations (pH): The pH of the all collected water samples was estimated *in situ* using pH model JWNWAY 3505.

Determination of cyanide levels: Free cyanide concentrations were determined in all collected water samples by using the photometric method using the Lovibond Microprocessor Multi direct Photometer with Reagent Test 0.01-0.5 mg L^{-1} CN as follows:

- Two milliliter of water sample was transferred to a clean vial (24 mm) of the apparatus and then 8 mL of deionized water was added to the vial, the cap was closed tightly
- The vial was placed in the sample chamber tell making sure that the marks were aligned, then we pressed (zero) key and the vial was removed from the sample chamber. Two level spoons No. 4 (white) cyanide-11 were added to the prepared water sample. The vial was tightly closed with the cap and was inverted several times to mix the contents. After that, tow level spoons No. 4 (white) cyanide-12 were added to the sample vial, then it was tightly closed with the cap and was inverted several times to re-mix all its contents. Finally, three drops of the same size from cyanide 13 were added to the vial by holding the reagent bottle vertically and squeezing slowly. The vial was tightly closed with the cap and was inverted several times to mix all its contents. The vial was placed in the sample chamber making sure that the marks were aligned and then it pressed the (test) key and wait for 10 min as a reaction period. After the reaction period has finished, the measurement was started automatically. The result was shown in the display in mg L^{-1} free cyanide

Statistical analysis of the data: Statistical analysis of data was carried out by using SPSS software version 19. One way ANOVA procedure and General Linear Models Procedure (GLM procedure) of SPSS software were used to analyses of variance. The results were presented as mean and standard errors (SE) for each variable. Significant differences between mean values were tested using Duncan's multiple range tests. Pearson Correlation estimated the correlation between pH and cyanide values. The p-value consider statistically significant when $p<0.05$.

RESULTS AND DISCUSSION

The analytical result of the number of positive cyanide samples and their percentage from the total number of the different collected water samples in each sampling site and also in relation to various water sources a re shown in Table 1, 2 and Fig. 2. Different sampling sites for each water source was individually represented to survey as much space as possible for each source at the period of the study, (Table 2, Fig. 1).

Table 1: Mean values, percentage of positive cyanide samples and pH values of all examined water sources

Water sources	Total No. of samples	No. of positive samples	Cyanide (Overall mean)	Ph mean values
Drinking water (site 1, 2 and 3)	28	16	0.0160 ± 0.004^{a}	7.6738 ± 0.05007^{a}
Irrigation water (site 4 and 5)	20	8	0.0160 ± 0.009^{a}	7.7543 ± 0.15602^{a}
Surface water (site 6, 7, 8 and 9)	28	17	0.0370 ± 0.017^{a}	7.6214 ± 0.05570^{a}
Wastewater (site 10 and11)	22	12	0.2195 ± 0.027^{b}	4.4450 ± 0.04500^{b}
Total	98	53		

^{a,b}superscript in the tables that do not share the same letter are significantly differ p<0.05

Table 2: Mean values (mg L) and percentage of positive cyanide samples in each sampling site in all examined water samples.

Water sources	Total No. of samples	No. of positive samples	Positive samples (%)	Cyanide level Mean±SE
Site 1	8	6	75.00	0.015 ± 0.010^{a}
Site 2	14	6	42.86	0.023 ± 0.011^{ab}
Site 3	6	4	66.67	0.010 ± 0.008^{a}
Site 4	14	6	42.86	0.015 ± 0.010^{a}
Site 5	6	2	33.33	0.017 ± 0.011^{ab}
Site 6	5	2	40.00	0.033 ± 0.011^{ab}
Site 7	10	6	60.00	0.049 ± 0.007^{b}
Site 8	3	2	66.67	0.040 ± 0.011^{ab}
Site 9	10	7	70.00	0.026 ± 0.007^{ab}
Site 10	16	8	50.00	0.396 ± 0.009^{c}
Site 11	6	4	66.67	0.043 ± 0.011^{ab}
Total	98	53	54.08	

^{a,b}superscript in the tables that do not share the same letter are significantly differ p<0.05, Site 1, 2, 3: Drinking water, Site 4, 5: Irrigation water, Site 6, 7, 8: Surface water Site 10, 11: Waste water

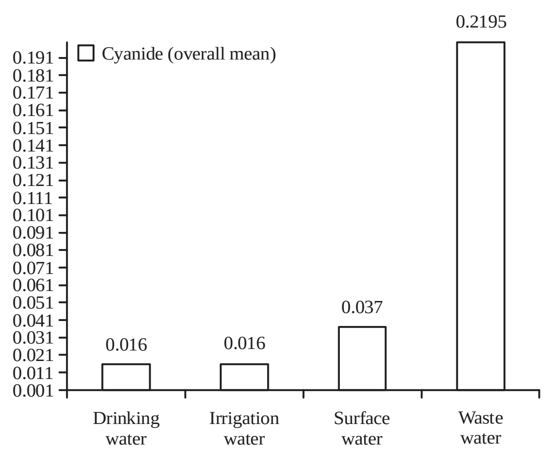

Fig. 2: Mean values of cyanide in the examined water sources

A total of 53 samples was positive for cyanide from 98 collected water samples as shown in Table 1. The overall mean values of cyanide in all tested drinking water samples, irrigation and surface water samples were within the permissible limits authorized by both USEPA (200 µg L^{-1}) and WHO (50 µg L^{-1}) according to USEPA[18] and WHO[19], respectively (Table 1, 2).

Surface water samples included 18 water samples from Al-Zennar canal and 10 pooled samples from the Nile River. The number of positive samples was higher in the Nile River (70 %) than Al-Zennar canal (55.55%). However, the mean values of cyanide in Al-Zennar canal (0.033 ± 0.011, 0.049 ± 0.007 and 0.040 ± 0.011 mg L^{-1} for site 6, 7 and 8, respectively) were higher than the Nile river (0.026 ± 0.007) as shown in Table 2. This can be explained by that Al-Zennar canal directly receives untreated wastewater collected from all over Assiut city, this makes the canal contains higher values of cyanides than the Nile River , in addition to the massive amount of water in Nile River come from its origin which plays a significant role in diluting the incoming pollutants.

In this study, 22 wastewater samples were collected from 2 wastewater treatment plant, at site 10, 11, respectively. The mean values of cyanide in all wastewater samples was 0.2195 ± 0.027 mg L^{-1} (Table 2). In addition, the mean cyanide value in wastewater site 10 (0.396 ± 0.009 mg L^{-1}), both exceeded the maximum permissible limit of cyanide concentrations imposed by the United States Environmental Protection Agency[18] (0.2 mg L^{-1}), WHO[19] (0.05 mg L^{-1}), The Council of the European Union[21] (0.07 mg L^{-1}).

From the statistical analysis of data, there was no significant difference between cyanide levels in the collected water samples from the same water source (Drinking, irrigation, surface) except for the wastewater (Table 1). Profoundly, the mean concentration of cyanide in the water samples collected at site 10 (WWTP 1) was significantly higher (p<0.05) than cyanide levels recorded at all water samples collected from other sites (Table 2). Concerning the significance of differences between different sites of sampling, cyanide level in site 7 (one of Al-Zennar canal sampling sites) was significantly higher than sites 1, 3, (drinking water sampling site) and site 4 (irrigation water sampling site), while site 7 was significantly lower than cyanide concentration that was shown in site 10 (WWTP1) (Table 2). This may attribute to the fact that Al-Zennar canal receives a large amount of wastewater from WWTP or may be contaminated by other sources of cyanide at the sampling time.

Regarding the different water sources, cyanide level in wastewater (0.2195$\pm$0.027) was higher than the surface water (0.037$\pm$0.017), irrigation water (0.016$\pm$0.009) and drinking water (0.016$\pm$0.004) (Table 1, Fig. 2). High level of cyanide in wastewater compared to other water sources is mainly attributed to receiving the WWTP of vast amounts of raw sewage, industrial effluent and municipal waste waters, which are the most significant source of cyanide in the environment[27].

The current data of the estimated pH values of different water sources showed a lower pH mean value of wastewater than all other water sources as shown in Table 1. Furthermore, wastewater pH mean value was significantly decrease from all other examined water sources (Table 1). Current results also showed a robust significant inverse negative correlation between water pH and cyanide ($p<0.0001$ and r-value was -0.953). This clearly means that cyanide increases at lower water pH and decreases at higher water pH. This result agreed with the finding of Meeussen *et al.*[28], who concluded that the increase in water acidity increases the dissociation of cyanide compounds into free cyanide in the water resources.

Furthermore, current results in line with Desai and Ramakrishna[11], who found that the stability of cyanide salts and metal-complexes cyanide in water bodies, as well as their potential hazards is pH dependent. Where, the metal-cyanide complexes are much less toxic than free cyanide, however, their dissociation releases free cyanide as well as the metal cation that can also be toxic. Furthermore, cyanide salts are highly soluble in water and thus readily dissolve to form free cyanide[11].

Free cyanide form estimated in this study is highly toxic to animals, human beings and all aquatic life when ingested. It reacts within a few hours to days with almost any other chemical they are exposed to, produce a wide variety of new compounds whose toxicity may be higher or less than the free cyanide itself[9]. Continuous ingestion of cyanide-contaminated water and crops even in low concentrations of both drinking and irrigation water, will possibly lead to chronic cyanide poisoning which will not be detected[29].

SIGNIFICANCE STATEMENT

This study is the first of its kind to evaluate the presence of cyanide in the waters of Assiut city. The presence of toxic cyanide in the water is a great danger to the health of humans and animals as well as the health of all living organisms and raise the alarm to re-evaluate the efficiency of water treatment plants and dealing with waste correctly and healthily and alert all the responsibility for this danger. It also opens the door for further research on the evaluation of the cumulative effect of drinking water contaminated with cyanide as well as the potential accumulation of this dangerous element in soil and agricultural plants. The study also shed light on more future studies to evaluate the efficiency of the treatment plants and drinking stations for this dangerous substance, as well as to limit the tolerability of the cyanide in reused water.

CONCLUSION

All examined water sources in the area of study including (drinking, irrigation, surface and wastewater) recorded positive samples of cyanide with different values and percentages. Cyanide values were the highest in wastewater samples and the lowest in both drinking and irrigation water samples. The presence of cyanide in 54.08% of the collected water samples and a higher concentration in waste waters exceeding the permissible limits is a real threat to the health of all living organisms, especially humans and animals. Therefore, the regular estimation of cyanide in different water sources and in biological materials that allow the assessment of risks resulting from environmental exposure to such hazard pollutant is highly recommended.

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