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# Toxicity, Physicochemical and Heavy Metals Analysis of Effluents Discharged from some Pharmaceutical Companies in Nigeria

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## ABSTRACT

Environmental impact assessment of ten (10) effluents from pharmaceutical industries in Lagos (4), Ogun (2) and Kwara (4) states in Nigeria was carried out in this study. Five freshwater tadpoles of fairly similar sizes were exposed to each of the effluent for 5 days. The physicochemical parameters, nutrient and heavy metals composition of the effluents were also evaluated using standard methods. After 5 days of observation, the mortality rate of the tadpoles ranged from 60% to 100% with all of Kwara effluents resulting in 100% mortality. Also, the positive control (tadpoles + fresh water) and negative control (tadpoles + laboratory tap water) had no mortality and 20% mortality rate respectively. The mean pH ranges (4.84 to 6.46), turbidity value (17.58 - 96.40 NTU), total dissolved solids (25.80 to 860.26 mg/L) and conductivity (28.50 to 205.0 $cm/s^{2)}$ of the effluents were outside the minimum acceptable range of the regulatory agencies. The dissolved nutrients, Chemical Oxygen Demand (COD), Dissolved Oxygen (DO) were also within regulatory limits except for nitrite and sulfide ion while the alkalinity of some samples exceeded the acceptable range. The organic compounds as well as the heavy metal concentrations were also within the stipulated limits, except Zinc and Magnesium.

**Keywords**: Pharmaceutical effluents, physico-chemical, toxicity, heavy metals, organic compounds, tadpoles.

## 1.0 INTRODUCTION

In the past few decades, the increase in human population and human activities (industrialization and urbanization) around the world has resulted in a corresponding increase in the production of wastewater containing harmful organic, inorganic compounds and heavy metals (Schweizer and Noblet, 2018). Developed countries such as Japan, France, Germany, the United Kingdom, and the United States account for two - third of the world's drug production and are major contributor to generatedpharmaceutical waste. Also, India and China are the sole producers of cheap natural drugs (WHO, 2014).

Approximately 70% of industrial trash in poor countries is often dumped untreated, contaminating nearby water bodies (Shrestha *et al.,* 2017; Ilomse*t al*., 2020). Numerous contaminants found in industrial wastes have been demonstrated to hinder the growth of various microorganisms, plants, and animals as well as acting as teratogenic, carcinogenic, and allergenic agents in humans (Chockalingam*et al.,* 2019). Furthermore, it is well recognized that the discharge of industrial effluent contains significant levels of suspended solids, high temperatures, heavy metals, and pH fluctuations. Industrial wastewater pollution destroys fisheries, aquatic life, and ecosystems in addition to seriously harming the health of nearby residents. It also affects the fertility of the soil.

Pharmaceutical-based pollutants are generated by the pharmaceutical industry in different operations (Gadipelly*et al.,* 2014). Various methods, such as chemical precipitation, cementing, floatation-flocculation, adsorption, biosorption, ion exchangers, and reverse osmosis (Zewail and Yousef, 2015; Sardella*et al.,* 2015; Zhu *et al.,* 2014) have been used to remove these hazardous wastes from the environment. Most pharmaceutical wastewater discharge is unregulated and suffused with pollutants. Many effluents from industries are poorly treated since the techniques used are selective. These techniques trap metallic ions of heavy metals in aqueous solutions leaving behind some defiant heavy metals in the pre-treated discharged effluent released into the environment. Most heavy metals that defy effluent pre-treatment techniques escape into surface and groundwater bodies where they accumulate and affect living resources – plants, aquatic organisms, and human health including a recently born baby, children, and adults (Xue*et al.,* 2017). The two sources of pharmaceutical compounds contaminating the human water meant for consumption are from (a) the production process of the pharmaceutical industry, and (b) the disposal mismanagement of widespread use of drugs and drug residues. These sources lead to the

escape of toxic metals in urban and agricultural wastewater. Generally, pharmaceutical compounds enter natural water through overflow or surface runoff from various non-point water sources of agricultural activities, and municipal and medical facilities (Yang *et al.,* 2016). Depending on the type of pharmaceutical and the extent of discharge into water bodies, the presence of a certain drug in a water source will vary from location to location.

Pharmaceutical presence and concentration in receiving water sources, which are the main entry point for drinking water, are influenced by natural attenuation, dilution, and the level of wastewater treatment used (WHO, 2021). The persistence of heavy metal contaminants in water bodies is due to their non-biodegradable property, which can prolong their polluting effects and increase their risk of possibly causing diseases or harm when exposed to biomes (Kanakaraju*et al.,* 2018). More so, toxic compounds are often detected in various water bodies including potable water predisposing humanity to potential health risks and adversely affecting the environment (Kanakaraju*et al.,* 2018).

Recalcitrant heavy metals discharged in effluents seep into drinking water as global scarcity of drinking water forces the population to depend on wells, streams, rivers, springs and lakes for their daily water needs. These toxic compounds are absorbed and accumulated in the human body via the food chain and other living resources, and cause health problems like cancers, nervous system and organ damage, and in severe cases death (Godwill*et al.,* 2019). Humans are exposed to toxic heavy metals in the environment through different routes including ingestion, inhalation and dermal absorption (Eqani*et al.,* 2016). Populaces in developing countries are more exposed to heavy metals. In addition, people can get in contact with heavy metals in their workplaces and environs. The growing demand for pharmaceuticals in Nigeria has led to an increase in pharmaceutical production companies resulting in more waste being generated, which are mostly recalcitrant, cytotoxic and genotoxic (Olaitan*et al*., 2014). The initial phase of a waterbody pollution control strategy involves a comprehensive evaluation and characterization of the wastewater discharges from manufacturing enterprises (AbuBakar*et al.,* 2020; Okereke *et al.,*2016). Although several researches have been carried out into the presence of pharmaceuticals in water bodies in Nigeria (Adesina and Felix, 2018; Ogunbanwo*et al*., 2022; Olarinmoye*et al*., 2022) which mostly were as a result of domestic sewage and hospital waste disposals, very few investigations have been carried out in Nigeria to assess the impact of discharged pharmaceutical waste effluents on the environment mostly limited to Lagos and Ogun states (Olaitan*et al*., 2014; Olusegun*et al*., 2021).

Therefore, samples of the physicochemical parameters of effluents from ten pharmaceutical industries located in the Nigerian states of Kwara, Ogun, and Lagos were gathered and analyzed to gain an understanding of the exposure routes and the effects that pharmaceutical pollutants have on man and his immediate environments.

## MATERIALS AND METHODS

### Study Area

The study area covered pharmaceutical companies in Lagos, Ogun, and Kwara States of Nigeria (Figure1, 2, 3). Thirty (30) pharmaceutical companies were identified in the three states based on their intensity of production, fame, and product quality. Seventeen pharmaceutical companies were identified in Lagos state with four permitting collections of effluent samples. Nine were identified in Ogun state but two permitted collections of effluent samples. Four were identified in Kwara state and all four approved collections of effluent samples.

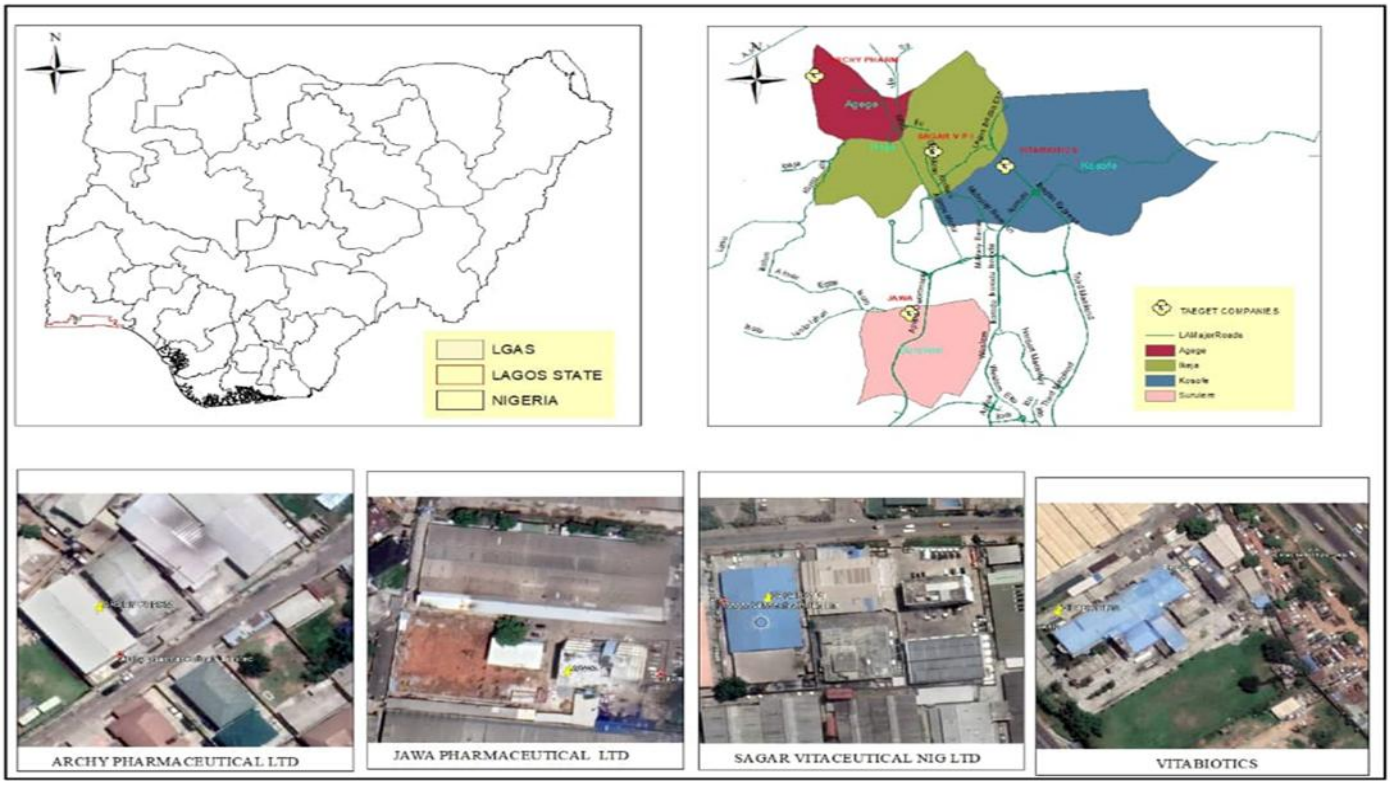



Figure 1: The locations of the pharmaceutical effluent samples taking in Lagos state of Nigeria.

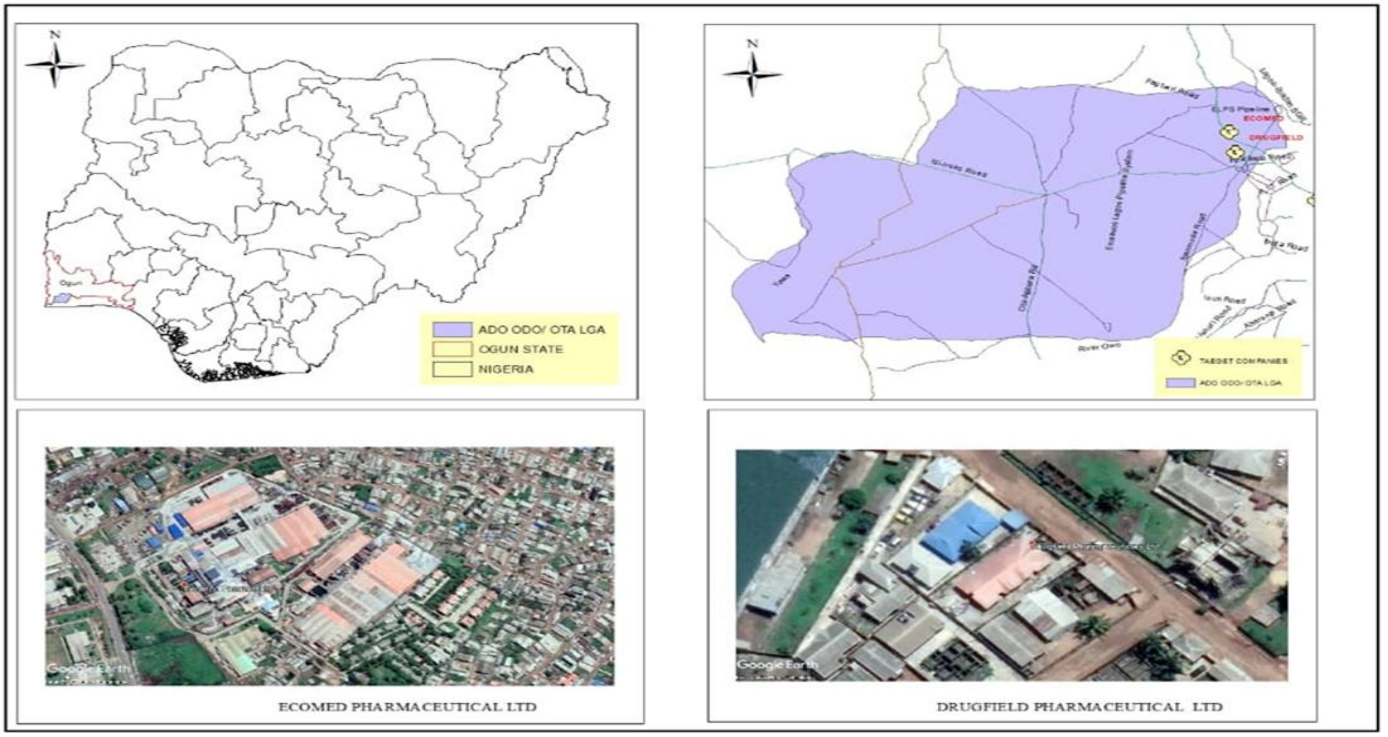



Figure 2: The locations of the pharmaceutical effluent samples taken in Ogun state, Nigeria.

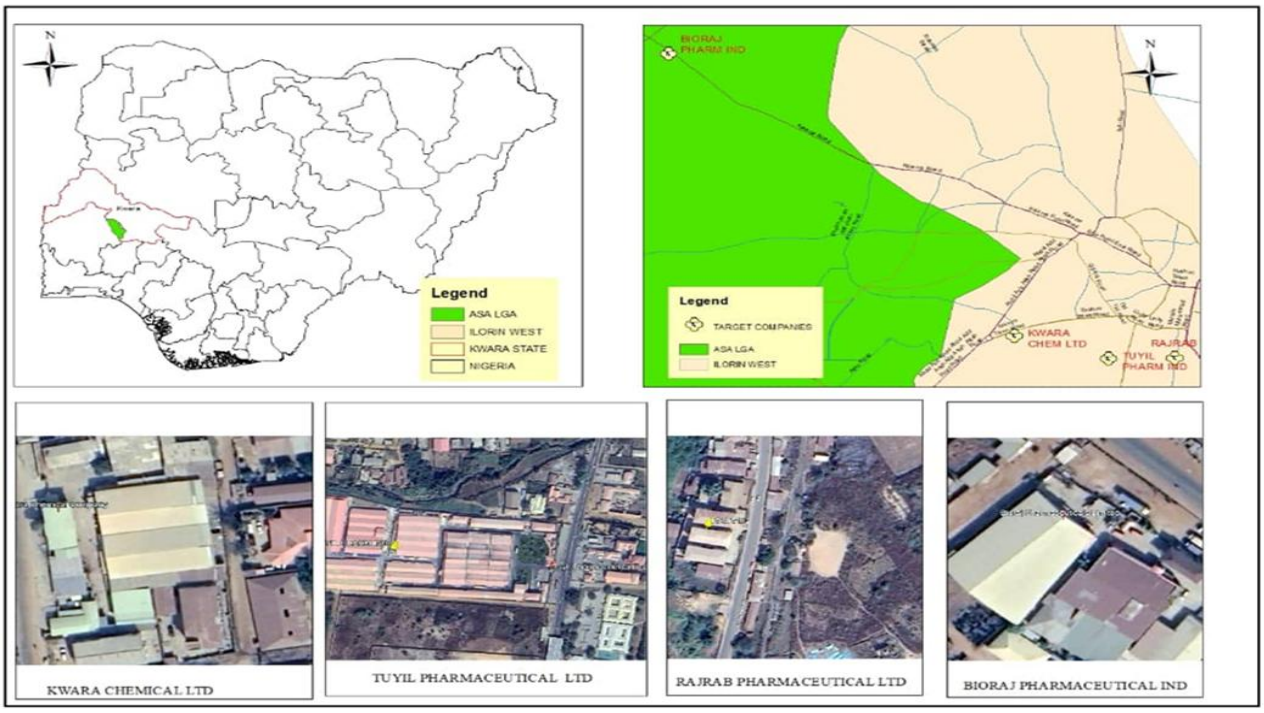



Figure 3: The locations of the pharmaceutical effluent samples taking in Kwara state, Nigeria.

**Collection of samples**

Ten pharmaceutical companies were visited for effluent sample collections for this study. Samples collected from these companies were designated with the letters of the alphabet (A to J). The samples were taken from companies discharge points twice, in raining season and dry season from October 2021 and February 2022.

The samples were collected into sterile, clean polythene bottles and rinsed with a 1:1 solution of nitric acid and distilled water after being pre-washed with detergent and distilled water. Three distinct sets of samples were gathered: one for each of the following analyses: toxicological, heavy metals, and physicochemical. 1.5 L of the samples from each sampling point was collected into the bottles with a label that indicates the date of collection, location, name of pharmaceutical company source, physical description of effluents, and collection surroundings. Afterward, these were placed in an ice chest and brought to the lab, where they were refrigerated at 4°C before being analyzed. (Barathi and Vasudevan, 2001). For this investigation, a total of thirty samples were gathered throughout different times of peak production across rainy and dry seasons.

The collected samples were analysed using standard methods (APHA, 2015) to identify the physicochemical, the concentration of each identified heavy metal, and the concentrations of nutrients/organic compounds present in each sampled source location.

Table 1: Collection periods for samples at various states.

| Collection Periods/Dates | Collection frequency | | |
|---|---|---|---|
| | Lagos state | Ogun state | Kwara state |
| 4$^{th}$October, 2021 | Four (4) samples per day in the morning | | |
| 4$^{th}$ October, 2021 | | Two (2) samples per day in the morning | |
| 7$^{th}$ October, 2021 | | | Four (4) samples per day in the morning |
| 7$^{th}$February, 2022 | Four (4) samples per day in the morning | | |
| 7$^{th}$ February, 2022 | | Two (2) samples per | |

| | | | |
|---|---|---|---|
| | | day in the morning | |
| 10th February, 2022 | | | Four (4) samples per day in the morning |

**Effluent Toxicity Test using Tadpoles**

Tadpoles of nearly the same sizes (5g) were used for the toxicity test. One hundred (100) tadpoles were collected from freshwater (stream) with the debris into a sterile container and transported to the laboratory. 200 ml of each effluent sample and five (5) active tadpoles were each placed in a sterile beaker and kept on the laboratory beach for 5 days. Two controls were set up and these were (control 1 contained 200 ml stream water and five (5) active tadpoles; control 2 contained 200 ml tap (borehole) water and five (5) active tadpoles. All were kept for 5 days in sterile beakers on the laboratory bench and observed daily for 5 days of mortality. The effects were characterized according to Park *et al.*(2016) and each day dead tadpole is removed. The toxicity was expressed by mortality rate, I% = (numbers of dead individual(s) /numbers of exposed tadpole individuals) X 100%.

**Physicochemical and Heavy Metal Analysis**

**A. Physical analysis of the samples**

The physical parameters of each sample were investigated according to the methods described by APHA (2015). These include true color using visual colorimetric methods, odor using the organoleptic method, pH using an electrometric digital pH meter, temperature using thermal gravimetric (thermometer), Turbidity using turbidimeter, Electrical Conductivity (EC) using electromagnetic induction (EC meter), Total Dissolved Solids (TDS) using a gravimetric method (electrical conductivity meter), Temperature using thermometer,

**B. Chemical analysis of the samples**

The Dissolve Oxygen was investigated using Azide modification colorimetric method while wet chemistry method was used to determine the Chemical Oxygen Demand (COD). Alkalinity and Hardness was assayed with titrimetric/colorimetric methods (APHA2015).

**C. Nutrient analysis of the samples**

The concentrations of nutrients such as Nitrate, Nitrite, Fluoride, Sulphate, Chloride, and Sulphide were done using colorimetric methods described by APHA, (2015).

**D. Heavy Metals Analysis**

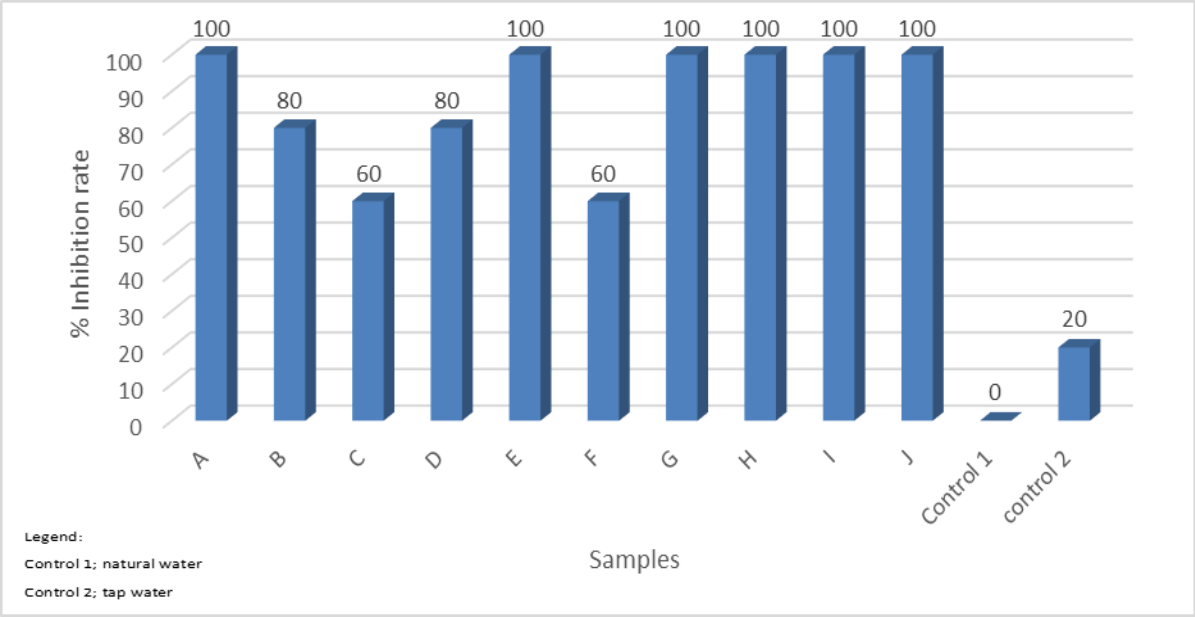
Traces of heavy metals such as Arsenic, Cadmium, Mercury, Manganese, Nickel, Iron, Zinc, Copper, Lead, and Chromium from the effluents were determined using A flame AAS (Atomic Absorption Spectrophotometer), after preparation of appropriate calibration standards according to APHA(2015) protocol.

**E. Determination of Organic Compounds**

Ammonia, mineral oil, pesticides, and total organic carbon were assayed using liquid chromatography. Polyaromatic hydrocarbon, Total trihalomethane, and 2,4,6 – trichlorophenol were also determined using gas chromatography. The detergent was determined using spectroscopic, Phenol compound was determined using spectrophotometric methods. All done according to APHA (2015) protocol.

**Results and Discussion**

The results of the mortality of tadpoles exposed to the sample effluents is shown in Figureure4.The effluents A, E, G, H, I and J had 100% mortality rate on all the five tadpoles at the end of 5 days while effluent B and D, as well as C and F had 80% and 60% mortality rate on four tadpoles and three tadpoles respectively at the end of 5 days. The positive control 1 which is the tadpoles + fresh water (stream water) collected from their natural habitat had 0% mortality rate at the end of 5 days on the five tadpoles while the negative control 2 which is tadpoles + tap water from the laboratory had 20% mortality rate on two tadpoles at the end of 5 days. The high mortality of the tadpoles when exposed to the effluents may be due to high COD of the effluents as well as possible hazardous chemicals. Low COD in samples C and F may have accounted for the survival of some tadpoles till the end of 5 days. More so, high pH and low temperature between $24^0$C -$26^0$C may account for the survival of some tadpoles. Kumari and Tripathi (2019) also observed high COD in pharmaceutical waste effluents and concluded that it might be as a result of chemical ingredients that were used in the preparation of drugs.




**Figureure4: Toxicity test of samples using Tadpoles**

Table 2 showed the physical characteristics of various effluent samples from ten pharmaceutical businesses (A to J) at the discharge points. Significant amounts of dissolved matter: turbidity, pH, conductivity, temperature, Total Dissolved Solids (TDS), true color, appearance, color and odor values are among the pollutants that make up the effluents' characteristics. Every physical features of the samples were within the NESREA/FEPA/WHO approved ranges with the exception of turbidity, The NESREA standard for electrical conductivity was not clearly defined.

**Turbidity**

Turbidity of the effluents ranged from 17.58 - 96.40NTU. Effluent F had the lowest turbidity of 17.58NTU while effluents H, I and A had the highest turbidity of 94.40, 94.30 and 92.50NTU respectively. The recorded values were observed to be significantly higher than regulatory agencies limits (5 to 10NTU).The high turbidity levels of the effluent samples may be due to the presence of some compounds /ingredients used in the production of drugs in the pharmaceutical industries. As a result, the turbidity of the effluent samples may hinder the light penetration that submerged aquatic vegetation requires, which could have an impact on primary production. It may also have an impact on organisms that are directly or indirectly dependent on aquatic primary production. It may also have an impact on fish gills'capacity to take in dissolved oxygen. One of the important factors that significantly affects the lotic and lentic systems' water quality is turbidity. High turbidity may seriously impair the streams, rivers, and lakes' aesthetic appeal,

which would lower the resources' recreational value. Different states have freshwater turbidity guidelines for streams and lakes because of the diverse effects that turbidity has on aquatic systems, the security of drinking water, and end users (SCDHEC, 2014).

**pH**

One of the key biotic features that acts as an index for pollution is the pH (Anjana and Janak, 2015). The pH of the effluents ranged from 4.84 - 6.69. Effluent I had the lowest pH value of 4.84 while effluents C and F had the highest pH values of 6.69 and 6.64 respectively. The pH values that were obtained from the samples fell somewhat short of the natural-7 pH of water, ranging from 4.84 to 6.46. The pH values obtained from some effluents discharged were lower than the maximum acceptable NESREA, WHO and FEPA limits of 6.0 to 9.0.All of the samples had somewhat acidic pH values, and the effluents' acidic nature has the potential to lower the pH of the receiving water bodies, which could upset essential elements including the water's hardness, alkalinity, and solubility of metals. The evidence presented by Wang *et al*. (2002) indicates that the pH levels have an impact on the metabolic processes of aquatic species. Therefore, the pH of these effluent may directly have adverse effects on the living resources when discharged into the surrounding environments. It may pose life threat to living resources and the aquatic habitats using the water for metabolic purposes. The pH of these effluent may directly have adverse effects on the living resources when discharged into the surrounding environments. It may pose life threat to living resources and the aquatic habitats using the water for metabolic purposes.

**Electrical Conductivity**

The conductivity of the effluents ranged from 28.50 – 205 us/cm$^2$. Effluent F and C had the lowest conductivity value of 28.50 and 50.0 us/cm$^2$ while effluents B and D, A and G had the highest conductivity values of 205, 172.5 and 128.4 us/cm$^2$ respectively. No specific standard limit was set by the regulatory agencies for electrical conductivity values. High conductivity levels show a high risk of corrosion and fall within the permitted range set by NESREA and FEPA, and WHO. This implies that the demineralized segment of the boiling and cooling water in the industry has low amounts of total dissolved solids. Water conductivity is a useful indication for determining the effluent's salinity (Idris *et al*., 2013).

**Temperature**

The temperature of the effluents ranged from 27.10 – 27.70$^0$C. Effluent C and F had the lowest temperature value of 27.10$^0$C while effluents B, H and I had the highest temperature values of

27.54, 27.60 and 27.70$^0$C respectively. One of the main elements influencing the aquatic environment is temperature (Wege*et al.,* 2012). The temperature of surface water contaminated by different effluents may decrease as a result of dilution (within the WHO permitted limit); this is advantageous for the survival of any local population of aquatic life species, but it also poses a serious health risk to living resources (Ogbu*et al*., 2016). Dilution of the effluents (within the WHO-permitted level) may cause surface water contaminated by various effluents to cool down, preserving the existence of any local populations of aquatic life species and posing a major health danger to living resources (Ogbu*et al*., 2016).

All temperature value derived from the samples were within NESREA (2009) permissible limits of (20 – 32$^0$C ambient temperature) for effluent discharges to surface water but did not meet the WHO standard (12 – 25$^0$C) and hence would harm the water quality using WHO standard; a temperature of 27.10$^0$C – 27.70$^0$C was maintained within the samples. This may pose danger and threat to aquatic organisms and habitats.

**Total Dissolved solids**

The total dissolved solids (TDS) of the effluents ranged from 25.80 – 860mg/L. Effluent F and E had the lowest TDS values of 25.80mg/L and 68.21mg/L respectively while effluents C, B and G had the highest TDS values of 127.45, 137.37 and 860.28mg/L respectively. The TDS values obtained from the effluent discharged were within the maximum acceptable NESREA, WHO and FEPA limits of <1200 to 2000 mg/L. The entire number of inorganic substances dissolved or suspended in water is measured as total dissolved solids (TDS) often known as suspended solids (Sonune*et al.,* 2015). Light intensity is influenced by the number of suspended solids in the water, which in turn affects turbidity and transparency (Kesharwani*et al.,* 2011).

**Colour and Odour**

The true colour of all the samples fell within NESREA (2009) permissible limits of 7.00 Hz. The colour of the samples was <0.01.All of the effluent samples had an offensive smell, and they did not fall below the unobjectionable NESREA (2009) permitted levels. The presence of odour in the effluent samples may be due to the composition and compounding of various drugs during production. Additionally, certain gases (ammonia and hydrogen sulphide) present in industrial pharmaceutical effluent can be hazardous, odorous, and offer an asphyxiation risk. This observation is consistent according to the report from Egwuonwu*et al*. (2012).

Table 2. The physicochemical properties of the pharmaceutical effluent samples

| Samples | Turbidity (NTU) | pH | Conductivity (us/cm) | Temp ($^0$ c) | Tds (mg/L) | True color (Hz) | Appearance | Color | Odor |
|---|---|---|---|---|---|---|---|---|---|
| A | 92.5 | 5.76 | 172.5 | 27.52 | 115.58 | <0.01 | Clear | Colorless | Objectionable |
| B | 45.33 | 5.16 | 205 | 27.54 | 137.37 | <0.01 | Clear | Colorless | Objectionable |
| C | 39.53 | 6.69 | 50.2 | 27.1 | 127.43 | <0.01 | Clear | Colorless | Objectionable |
| D | 45.33 | 5.16 | 205 | 27.53 | 102.91 | <0.01 | Clear | Colorless | Objectionable |
| E | 45.33 | 5.16 | 101.8 | 27.5 | 68.21 | <0.01 | Clear | Colorless | Objectionable |
| F | 17.58 | 6.46 | 38.5 | 27.1 | 25.8 | <0.01 | Clear | Colorless | Objectionable |
| G | 45.52 | 5.18 | 128.4 | 27.53 | 860.28 | <0.01 | Clear | Colorless | Objectionable |
| H | 94.4 | 5.76 | 100.5 | 27.6 | 155.48 | <0.01 | Clear | Colorless | Objectionable |
| I | 94.3 | 4.84 | 100 | 27.7 | 112.56 | <0.01 | Clear | Colorless | Objectionable |
| J | 30.54 | 5.18 | 103.8 | 27.3 | 125.58 | <0.01 | Clear | Colorless | Objectionable |
| NESREA limits | 10 | 6.00-9.00 | NS | Ambient | 2000 | 7.00 | Clear | Colorless | Unobjectionable |
| FEPA limits | NS | 6.0-9.0 | NS | < 40 | 2000 | NS | NS | NS | NS |
| WHO limits | 5 | 6.0-9.5 | NS | < 36 | < 1200 | NS | NS | NS | NS |
| USEPA limits | NS | 6.5-8.5 | NS | NS | 500 | NS | NS | NS | NS |
| RANGES | 17.58-96.40 | 4.84-6.69 | 28.50-205 | 27.10-27.70 | 25.80-860.28 | <0.00 - 0.01 | Clear | Colorless | Objectionable |
| MEAN | 54.25 | 5.33 | 134.57 | 27.53 | 183.12 | <0.01 | Clear | Colorless | Objectionable |
| SDEV | ±28.96 | ±0.31 | ±55.88 | ±0.09 | ±240.77 | <0.01 ± 0.00 | Clear | Colorless | Objectionable |

**LEGEND: NS = Not Stated**

Table 3 showed the chemical analysis of the effluent samples from the ten pharmaceutical companies (A to J). The total hardness ranged from 25.0 – 272.25 mg/L with Effluent F having the lowest total hardness (25 mg/L) while the highest total hardness was recorded in effluent C (272.25mg/L). The regulatory agencies limits were however not stated. Given that the receiving water's hardness ranged from 25 to 272.25mg/L, it can be said that the water is extremely hard and would need chemical treatment (McGowan, 2000).

The free chloride of the effluents was found to be less than 0.0001mg/L. There were no specified limits for free chloride by the regulatory agencies. With this result, the values fell within the set

regulatory standard. It was also observed that the total alkalinity of the effluent ranged from 40 to 380mg/L. Effluent F had lowest alkalinity of 40mg/L, while Effluent H had the highest alkalinity of 380mg/L. Effluent A, E, F, and G fell within the regulatory standard (200mg/L). The dissolved oxygen (DO) of the effluent ranged from 1.17 to 1.80mg/L. Effluent G, H, I and J had 1.17mg/L, effluent A, B, D and E had 1.18mg/L while effluent C and F had 1.80mg/L of the DO. All the effluent fell within the regulatory limit of 2.00mg/L. The low DO values may have contributed to the high alkalinity values, which ranged from40 to 380 mg/L (Olaitan*et al.,* 2014**).** Furthermore, the COD values ranged from 0.07 - 4.79 mg/L which were all within the stipulated limits by NESREA for surface water (80mg/L). Both the chemical oxygen demand (COD) and the Biological Oxygen Demand (BOD), which are functions of DO, are helpful factors in the investigation of water quality. COD was very low which is normal for such pharmaceutical industries. Hence, there would be no effect of effluent on the surrounding environment (Eqwuonwu*et al*., 2012).

Table 3: Chemical Analysis of the samplesin Ranges, Means and Standard Deviation

| **SAMPLES** | **Total Hardness (mg/l)** | **Free Chlorine (mg/l)** | **Total Alkalinity (mg/l)** | **Dissolved Oxygen (mg/l)** | **COD (mg/l)** |
|---|---|---|---|---|---|
| A | 163.75 | <0.0001 | 180 | 1.18 | 0.36 |
| B | 225.00 | <0.0001 | 215 | 1.18 | 0.15 |
| C | 272.25 | <0.0001 | 226 | 1.80 | 0.12 |
| D | 165.50 | <0.0001 | 250 | 1.18 | 0.18 |
| E | 94.25 | <0.0001 | 50 | 1.18 | 0.18 |
| F | 25.00 | <0.0001 | 40 | 1.80 | 0.07 |
| G | 163.35 | <0.0001 | 180 | 1.17 | 4.76 |
| H | 263.75 | <0.0001 | 380 | 1.17 | 3.99 |
| I | 163.75 | <0.0001 | 210 | 1.17 | 3.74 |
| J | 113.75 | <0.0001 | 225 | 1.17 | 4.79 |
| NESREA LIMITS | NS | NS | 200.00 | 2.00 | 80.00 |
| FEPA LIMITS | NS | NS | NS | NS | NS |
| WHO LIMITS | NS | NS | NS | NS | NS |
| USEPA LIMITS | NS | NS | NS | NS | NS |
| RANGES | 25.0- 272.25 | <0.0001 | 40.0 -380 | 1.17-1.80 | 0.07-4.79 |
| MEAN | 164.04 | <0.0001 | 195.6 | 1.18 | 1.83 |
| SDEV | ±76.16 | ±0.00 | ±97.32 | ±0.00 | ±2.16 |

LEGEND: NS = Not Stated

Table 4 showed the concentration of nutrient in the effluent samples from ten pharmaceutical companies (A to J).

The Chloride ions of the effluents ranged from 10.64 to 65.60mg/L. Effluent E, J, A and I had the lowest value of 10.64, 12.80 and 13.80mg/L respectively while effluent B and C had the highest value of 56.74 and 65.60mg/L. All the effluents fell within the ranges of regulatory limits of 250 to 600mg/L, Also the fluoride ions content of the effluents was found to be less than 0.0001mg/L. This fell within the regulatory limits of 1.50mg/L. Also, the nitrate ions content of the effluents ranged from 1.17 to 3.41mg/L. Effluent G and J had the lowest value of 1.20 and 1.30mg/L respectively while effluent C and E had the highest value of 3.41 and 3.39mg/L. All the effluent fell within the ranges of regulatory limits of 10 to 50mg/L.

The nitrite ions content of the effluents ranged from 0.08 to 2.47mg/L. Effluent C and D had the lowest value of 0.12 and 0.17mg/L respectively while effluent F had the highest value of 2.47mg/L. Some of the effluent fell within the ranges of regulatory limits of 0.20mg/L while some effluents were above the regulatory limit. The sulphate values of the effluents ranged from 1.73 to 49.33mg/L. Effluent C and J had the lowest value of 1.73 and 4.57mg/L respectively while effluent B had the highest value of 49.33mg/L. All the effluent fell within the ranges of regulatory limits of 100 to 500mg/L.Sulphide ions of the effluent was found to range from 2.15 to 8.15mg/L. Effluent E and D had the lowest value of 2.15 and 2.45mg/L respectively while effluent A and C had the highest value of 6.75mg/L and 8.15mg/L.

All the effluent did not fall within the range of regulatory limits of 0.05mg/L. Sewage discharges, chemical industry effluent discharges, salt deposits, and other sources are responsible for the occurrence of chloride in natural water. Surface and groundwater contamination in the area could result from any of these sources (Boghra*et al.*, 2011). Both plant and animal growth and metabolism depend on trace amounts of sulphates and nitrates. However, these minerals can be hazardous in excess since they can generate an algal bloom and lower the amount of oxygen in the water, both of which can kill aquatic life (Ansar and Khad, 2005).

Also, in Table 5 the organic compound components of the effluent samples from the ten pharmaceutical companies sampled is presented.It was observed that the ammonia content of all the effluents was less than 0.0001 mg/L which was below the regulatory agencies limits of 0.05mg/L. The Mineral oil content of all the effluents ranged from 0.0024 to 0.0082mg/L. Effluent F, E and C had the lowest mineral oil value of 0.0024, 0.0027 and 0,0030mg/L respectively while effluent J, G, and H had the highest value of 0.0068mg/L, 0.0076mg/L and

0.0082mg/L. Only the effluents A, B, C, D E,F and I had values that fell within the regulatory agency limit. It was also observed that the Phenol content, pesticides, aromatic hydrocarbon contents of the effluents were found to be within accepted limits while the detergent contents of all the effluents ranged from 0.065mg/L to 0.085mg/L. Pesticides, detergent, total organic carbon, aromatic hydrocarbon, and trihalomethane were all within the limit standard set by NESREA. They do not have effect on the aquatic organisms when discharged into the stream and river. High concentrations of these will have toxic effect on the aquatic resources (Ariful-Islam*et al.*, 2020;Seujpriya, 2022; Pozzer*et al*., 2022).

Table 4: Concentration of Nutrient in the Samples in Ranges, Means and Standard Deviation

| SAMPLES | Chloride (mg/L) | Fluoride (mg/L) | Nitrate (mg/L) | Nitrite (mg/L) | Sulphate (mg/L) | Sulphide (mg/L) |
|---|---|---|---|---|---|---|
| **A** | 13.80 | <0.0001 | 1.6 | 0.48 | 5.77 | 6.75 |
| **B** | 56.74 | <0.0001 | 1.17 | 0.19 | 49.33 | 3.00 |
| **C** | 65.60 | <0.0001 | 3.41 | 0.12 | 1.73 | 8.15 |
| **D** | 14.18 | <0.0001 | 1.56 | 0.17 | 19.67 | 2.45 |
| **E** | 10.64 | <0.0001 | 3.39 | 0.08 | 19.33 | 2.15 |
| **F** | 27.30 | <0.0001 | 2.61 | 2.47 | 4.3 | 4.45 |
| **G** | 53.50 | <0.0001 | 1.2 | 0.52 | 5.77 | 6.25 |
| **H** | 23.80 | <0.0001 | 1.3 | 0.28 | 4.77 | 4.75 |
| **I** | 13.80 | <0.0001 | 1.2 | 0.52 | 6.77 | 6.55 |
| **J** | 12.80 | <0.0001 | 1.3 | 0.28 | 4.57 | 6.25 |
| **NESREA LIMITS** | NS | 1.50 | 20.00 | 0.20 | 100.00 | 0.05 |
| **FEPA LIMITS** | 600.00 | NS | 20.00 | NS | 500.00 | NS |
| **WHO LIMITS** | 250.00 | NS | 50.00 | NS | 500.00 | NS |
| **USEPA LIMITS** | 250.00 | NS | 10.00 | NS | 250.00 | NS |
| **RANGES** | 10.64 – 65.60 | <0.0001 | 1.17-3.41 | 0.08-2.47 | 1.73-49.33 | 3.15-8.15 |
| **MEAN** | 29.22 | <0.0001 | 87 | 0.510 | 12.2 | 5.08 |
| **SDEV** | ±21.14 | ±0.00 | ±0.91 | ±0.71 | ±14.46 | ±2.04 |

LEGEND: NS = Not Stated

Table 5: Organic Compound in Effluent Samples in Ranges, Means and Standard Deviations

| SAMPLES | Ammonia (mg/L) | Mineral Oil (mg/L) | Phenol Compounds (mg/L) | Pesticides (mg/L) | Detergent (mg/L) | AHC (mg/L) | TOC (mg/L) | TTM (mg/L) |
|---|---|---|---|---|---|---|---|---|
| A | <0.0001 | 0.0064 | <0.0001 | <0.0001 | 0.085 | <0.0001 | <0.0001 | <0.0001 |
| B | <0.0001 | 0.0064 | <0.0001 | <0.0001 | 0.083 | <0.0001 | <0.0001 | <0.0001 |
| C | <0.0001 | 0.003 | <0.0001 | <0.0001 | 0.082 | <0.0001 | <0.0001 | <0.0001 |
| D | <0.0001 | 0.0042 | <0.0001 | <0.0001 | 0.082 | <0.0001 | <0.0001 | <0.0001 |
| E | <0.0001 | 0.0027 | <0.0001 | <0.0001 | 0.08 | <0.0001 | <0.0001 | <0.0001 |
| F | <0.0001 | 0.0024 | <0.0001 | <0.0001 | 0.081 | <0.0001 | <0.0001 | <0.0001 |
| G | <0.0001 | 0.0076 | <0.0001 | <0.0001 | 0.085 | <0.0001 | <0.0001 | <0.0001 |
| H | <0.0001 | 0.0082 | <0.0001 | <0.0001 | 0.076 | <0.0001 | <0.0001 | <0.0001 |
| I | <0.0001 | 0.0059 | <0.0001 | <0.0001 | 0.074 | <0.0001 | <0.0001 | <0.0001 |
| J | <0.0001 | 0.0068 | <0.0001 | <0.0001 | 0.065 | <0.0001 | <0.0001 | <0.0001 |
| NESREA LIMITS | 0.05 | 0.003 | 0.01 | 0.01 | NA | 5.00 | 0.001 | 0.002 |
| FEPA LIMITS | NS | NS | NS | NS | NS | NS | NS | NS |
| WHO LIMITS | NS | NS | NS | NS | NS | NS | NS | NS |
| USEPA LIMITS | NS | NS | NS | NS | NS | NS | NS | NS |
| RANGES | <0.0001 | 0.0024-0.0076 | <0.0001 | <0.0001 | 0.065-0.085 | <0.0001 | <0.0001 | <0.0001 |
| MEAN | <0.0001 | 0.005 | <0.0001 | <0.0001 | 0.079 | <0.0001 | <0.0001 | <0.0001 |
| SDEV | ±0.00 | 0±0.00 | ±0.00 | ±0.00 | ±0.006 | ±0.00 | ±0.00 | ±0.00 |

Legend: NS: Not Stated**,** AHC= Aromatic Hydrocarbons, TOC= Total Organic Carbon, THM= Trihalomethanes

The concentrations (mg/L) of Heavy Metals in the effluent samples from ten pharmaceutical companies is shown in Table 6. The Lead concentration values of the effluents ranged from 0.00mg/L to 0.18mg/L with the concentrations in effluent H and J exceeding the maximum regulatory agency limit (0.08 mg/L).The magnesium ions values of the effluents ranged from 12.52mg/L to 72.93mg/L and they all exceeded the maximum regulatory limits of 0.20mg/L. Also, the heavy metal zinc exceeded the regulatory limits; ranging from 75mg/Lto 185.03mg/L while some of the effluents had copper concentrations exceeding the limits of 0.01 mg/L. High zinc concentration may cause bitter taste and undesirable effect to aquatic organisms. Aquatic resources and other creatures are harmful to heavy metals only when they are released into the environment in excessive concentrations (Hama Azi*z et al.,* 2023).

Iron/Manganese/Arsenic/Cadmium/Mercury/Chromium and Nickel of the effluents were below 0.0001mg/L which were all within the maximum regulatory limits of 0.03 to 20.00mg/L, 0.20mg/L, <1mg/L, 0.003mg/L, <1mg/L, 0.01 to 0.5mg/L and 0.01 to 0.05mg/L respectively. While fishes rely on essential metals to grow and use feed, extremely high concentrations tend to disrupt their normal physiological and ecological processes in the aquatic environment (Ediagbonya*et al.*, 2022). It was observed that the average levels of most of the parameters of all the pharmaceutical industrial effluents at their point of discharge were lower than the discharge specifications set by the regulatory bodies (WHO, 2002, Osaigbovo and Orhue, 2006, FEPA, 1991).

Table 6: Heavy Metals Concentrations (mg/L) in the samples in Ranges, Means and Standard Deviation

| **SAMPLES** | **Lead (mg/L)** | **Iron (mg/L)** | **Magnesium (mg/L)** | **Manganese (mg/L)** | **Zinc (mg/L)** | **Copper (mg/L)** | **Arsenic (mg/L)** | **Cadmium (mg/L)** | **Mercury (mg/L)** | **Chromium (mg/L)** | **Nickel (mg/L)** |
|---|---|---|---|---|---|---|---|---|---|---|---|
| A | 0.00 | <0.0001 | 48.62 | <0.0001 | 130.76 | 0.06 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| B | 0.00 | <0.0001 | 72.93 | <0.0001 | 127.49 | 0.00 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| C | 0.00 | <0.0001 | 68.70 | <0.0001 | 185.03 | 0.04 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| D | 0.00 | <0.0001 | 48.62 | <0.0001 | 127.49 | 0.05 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| E | 0.00 | <0.0001 | 28.32 | <0.0001 | 75.19 | 0.01 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| F | 0.00 | <0.0001 | 12.52 | <0.0001 | 33.67 | 0.00 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| G | 0.00 | <0.0001 | 47.62 | <0.0001 | 125.76 | 0.01 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| H | 0.18 | <0.0001 | 44.32 | <0.0001 | 124.72 | 0.01 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| I | 0.00 | <0.0001 | 58.63 | <0.0001 | 150.76 | 0.01 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| J | 0.16 | <0.0001 | 44.62 | <0.0001 | 120.76 | 0.06 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| NESREA LIMITS | 0.01 | 0.30 | 0.20 | 0.20 | 3.00 | 1.00 | 0.01 | 0.003 | 0.01 | 0.01 | 0.01 |
| FEPA LIMITS | < 1 | 20.00 | NS | 5.00 | < 1 | < 1 | < 1 | < 1 | < 1 | 0.05 | < 1 |

| | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| WHO LIMITS | 0.01 | 15.00 | NS | 0.05 | < 1 | NS | < 1 | 0.03 | < 1 | NS | 0.02 |
| USEPA LIMITS | 0.003 | 0.03 | NS | 0.05 | 0.12 | 0.01 | < 1 | 0.02 | < 1 | NS | 0.05 |
| RANGES | 0.00-0.18 | <0.0001 | 12.52-72.93 | <0.0001 | 33.67-185.03 | 0.00-0.06 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| MEAN | 0.03 | <0.0001 | 47.49 | <0.0001 | 120.16 | 0.02 | <0.0001 | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| SDEV | ±0.07 | ±0.00 | ±17.74 | ±0.00 | ±40.72 | ±0.03 | ±0.00 | ±0.00 | ±0.00 | ±0.00 | ±0.00 |

LEGEND: NS =Not Stated

## Conclusion

The results of this study show that the majority of the parameters were within the acceptable regulatory ranges thus showing that the effluents were treated before discharge.Such effluents can be used for irrigation purposes at low levels it significantly impacting the ecosystem in a way that affects human, animal, or plant life.It is recommended that all the pharmaceutical industries that have not been treating their effluent discharged properly, should treat it properly before release to surface waters and the environment.

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