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Ecotoxicity potentials of residual paracetamol and personal care products (PCPs) in household waste in the University of Port Harcourt, Port Harcourt, Nigeria:

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Abstract

This study evaluated the relative amount of Pharmaceutical and Personal Care Products (PPCPs) and paracetamol residues contained in household waste at the University of Port Harcourt, Port Harcourt, Nigeria. Residual paracetamol was analysed using a 6405 UV/Vis Spectrophotometer at the maximum wavelength (245nm) in 0.1N HCL. Student hostels had a mean paracetamol concentration of 0.67 mg/kg while the staff quarters recorded a mean concentration of 0.66 mg/kg. Solid waste analysis recorded 0.9 kg/capita/day and 1.1 kg/capita/day for the student hostel and staff quarters respectively. The student's hostel recorded a mean percentage PPCPs value of 52.9% which was higher than that obtained in the staff quarters (45.9%) indicating that students disposed of more drugs in their waste than does the staff. The study showed that considerable amounts of PPCPs are disposed of in household waste at the study location and household waste is thus a viable pathway of PPCPs into the environment. Such practices could lead to the occurrence of the active ingredients of PPCPs in the environment with the potential to contaminate surface and ground water posing serious risk to human and ecologic health. It was recommended that appropriate return channels be established specially as a route for disposing of unused or expired drugs. Such routes should encourage stewardship of drugs by manufacturers from cradle to grave in an integrated green pharmacy approach.

Keywords: *Household waste, Pharmaceuticals and personal care products (PPCPs), residual paracetamol*

1. Introduction

Pharmaceuticals and Personal Care Products (PPCPs) are produced and used in increasingly large amount each year wherever man lives. Concerns about the fate and effects of these compounds in the environment has become topical among the scientific community. As opposed to the priority pollutants whose discharge into the environment are now more surreptitious due to regulatory activities PPCPs have enjoyed and are still enjoying unlimited and unregulated discharge into the environment with little knowledge of their potential harm to human and ecological health. This has warranted that detailed investigations be carried out on these contaminants using paradigm methodologies by scientists from different disciplines. Results from such investigations will reveal possible trends in occurrence, fate, exposure and potential physiological effects on ecological and human health. More available data can then form the basis for regulatory or policy decisions on this emerging environmental problem. Moreover,

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early identification and investigation of a potential environmental problem before it worsens is critical to protecting human and ecological health and also necessary to determine whether the issue is of concern or not so that limited resources can be redirected to other more pressing issues (Daughton and Ternes, 1999).

PPCPs comprise animal and human medicines, diagnostic agents, nutraceuticals (antibiotics, aphrodisiacssexual enhancement drugs and personal care products such as cosmetics, fragrances, detergents, feminine sanitary products , disinfectants, insect repellants (Pfleger 1999). Their sources include human activity, residues from pharmaceutical manufacturing, residues from hospitals, illicit drugs such as cocaine and methamphetamine, veterinary drug use especially antibiotics and steroids, agro-allied business etc. (Daughton and Ternes, 1999). Their discovery in environmental samples especially in Europe and America has given rise to concerns over their pathways into the environment and the extent of environmental occurrence, transport, fate and possible physiologic effects on ecological and human health. Comprehensive literature on this emerging environmental problem is still limited. Data available is mostly from Europe and America where PPCPs have been reported to occur at very low levels parts per trillion and parts per billion (ppt and ppb) in various environmental media such as soils, (Aga and Thurman, 2001; Rabolle and Spliid, 2000), biosolids (Xia *et al*., 2005), surface waters, under-ground water and even drinking water (Heberer, 2002; Heberer and Stan, 1997; Kolpin*et al*., 2002; Haggard *et al*., 2006,Xia *et al.*, 2005), landfill leachates (Eckel *et al*., 1993; Holm *et al*. 1995; Ahel and Jelicic, 2001) and household waste (Jonathan and Nikolaos, 2005). PPCPs are said to have high affinity for sediments (Giger *et al*., 2003 and Zweiner *et al.*, 2000) in which PPCPs were recovered about 50% more in sediments than in the overlaying surface water.

The primary source and pathways of PPCPs into the environment stems from their use and disposal by people all over the world and household waste constitute one of the primary routes for domestic disposal of PPCPs (Jonathan and Nikolaos, 2005). What is more worrisome is people are yet to accept the fact that chemical pollution can occur from consumption actions of individuals (Daughton, (2003). As a result, household chemicals (e.g. detergents, deodorizers, degreasers, disinfectants etc), pharmaceuticals (e.g. caffeine, hormones, steroids, antibiotics, etc) and other personal care products (PCPs)(e.g., fragrances and cosmetics) are used in large quantities and washed, excreted and flushed down sinks and toilets and their packaging materials disposed along household waste without a second thought. It has been reported that between 50 to 90% of some drugs ingested are excreted as its active compound or as a conjugate metabolite (Jonathan and Nikolaos, 2005) into the toilets or directly into water ways as practice in most poor nations like Nigeria. In developed nations, consumers are encouraged to return expired drugs or unused drugs to pharmacies or flushed them down the toilet.

A wide range of pharmaceuticals has been found in fresh and marine waters, and it has recently been shown that even in small quantities, some of these compounds have the potential to cause harm to aquatic life. The primary pathway into the environment is the use and disposal of medicines; although much of the research in this area currently focuses on the removal of pharmaceuticals during sewage treatment processes, disposal via household waste might be a significant pathway requiring further research. Jonathan and Nikolaos, (2005) demonstrated that disposal of unused pharmaceuticals, either by household waste or via the sink or toilet, may be a prominent route that requires greater attention. The present research investigated the relative composition of PPCPs and residual paracetamol in household waste in two locations at the

University of Port Harcourt, Nigeria to determine if household waste is a significant pathway of PPCPs into the environment.

Materials and Methods

Study Area

The investigation was carried out at University of Port Harcourt, Nigerian in Rivers State between these coordinate $4^0$54'01''N, $6^0$55'30"E, Fig. 1. This study was carried out in one of the university staff quarters (Gambia-ama) and Block D of the female hostel (Delta Park) in University of Port Harcourt which has five wings (A-F). These Locations were chosen because they represent typical urban dwelling in Nigeria, reflecting government quarters and hostel accommodation. Ten houses of the staff quarters were randomly sampled to reduce bias and also the female hostel of the University was surveyed to determine their possible contribution of PPCPs into the environment.

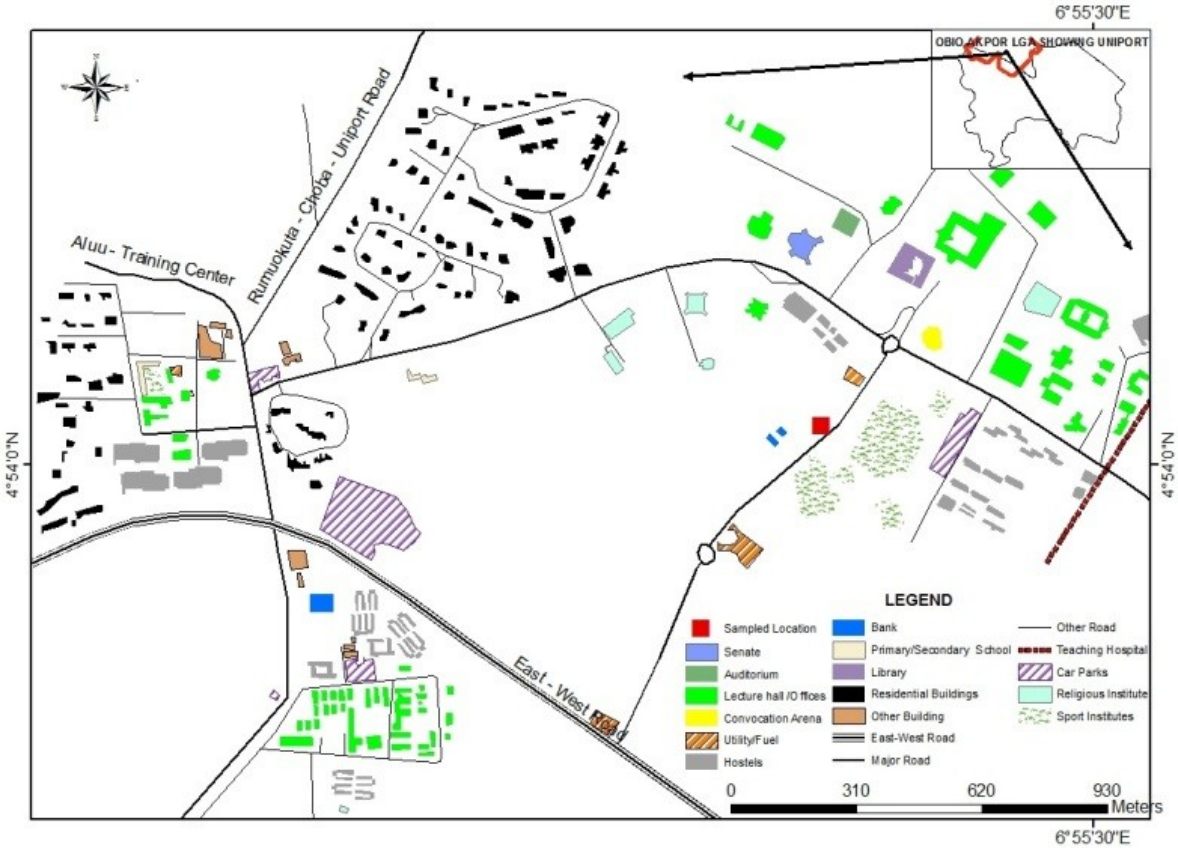

Figure 1: Map of University of Port Harcourt showing study site in red box

Methods

Collection and sorting of samples

This study was carried out in one of the University staff quarters (Gambia-Ama) and Block D of the female hostel (Delta Park) in the University of Port Harcourt which has five wings (A-F). These Locations were chosen because they represent typical urban dwelling in Nigeria, reflecting government quarters and hostel accommodation. Houses were numbered and ten houses of the staff quarters were randomly sampled to reduce bias and also the female hostel of the University was surveyed to determine their possible contribution of PPCPs especially paracetamol into the environment. Bags for perishable and non-perishable wastes were distributed to the randomly selected houses and the hostel weekly for one month for the disposal of their wastes. These bags were retrieved weekly, weighed and sorted manually according to ASTM D5231 - 92(2008) standard method which involves thamol'e direct sampling of solid waste from specific sources. Household wastes were sorted out into two different categories of solid waste and PPCPs. The PPCPs were weighed again to determine its composition in the solid waste and the paracetamol sachets found were taken to the laboratory where they were analyzed to determine the amount of active ingredient left in them.

Preparation of standard curve for paracetamol

A 50 mg quantity of standard paracetamol was dissolved in about 10 ml of absolute alcohol (Sigma, USA Analytical grade) and made up to 50 ml using 0.1 N HCL (Analytical grade Hydrochloric acid) to give a 100 mg stock solution (1mg/ml). From this stock solution, aliquots of 0.1-0.5 ml were measured and each made up to 50 ml using 0.1 N HCL. Their respective absorbances were determined at 245 nm using a 6405 UV/Vis spectrophotometer (Jenway, USA).

Determination of paracetamol content in samples

Aliquots of 95 % ethanol was used to wash off the paracetamol from the sachets. Intact tablets were pulverized, dissolved in ethanol and filtered. The entire dissolutions were made up to 100ml using ethanol. A 5 ml volume collected from the stock solution of sample dissolutions was made up to 100 ml using 0.1 N HCl. The content of paracetamol in this solution was determined spectrophotometrically at 245 nm.

Results

Solid Waste Generated per capita

The quantity of solid waste generated in the staff quarters and student's hostel is presented in figures 2 and 3 respectively. The mean quantity of waste generated ranged from 2.775 kg/day in house 2 to 8.550 kg/day in house 8 in staff quarters and from 3.075 kg in wing A to 3.975 kg in wing F in the student hostels. House 8 generated the highest average quantity of waste in weeks one to three while house three recorded the highest in week four. House 2 recorded the least quantity of waste in weeks one and four, house 3 recorded the least in week three while house four recorded the least quantity of waste in week two Fig 2. In the female hostel, wing C recorded the highest quantity of waste in week one, wing F recorded the highest in week two and four while wing E recorded the highest quantity of waste in week three Fig. 3.

Relative amounts of PPCPs in staff quarters and students hostel

The percentage composition of PPCPs in the waste from the staff quarters and students hostel is presented

in figures 4 and 5 respectively. The percentage PPCPs in the waste from the staff quarters ranged from 38.94% in house 6 to 52.26% in house 1 while that of student's hostel ranged from 39.69% in wing A to 65.29% in wing C.

Residual concentration of paracetamol in waste

The residual concentration of paracetamol contained in waste from staff quarters and student's hostel showed that the concentration of the residual paracetamol contained in waste from student's hostel was 0.6702 mg/kg of waste while that contained in staff quarters was 0.6569 mg/kg of waste.

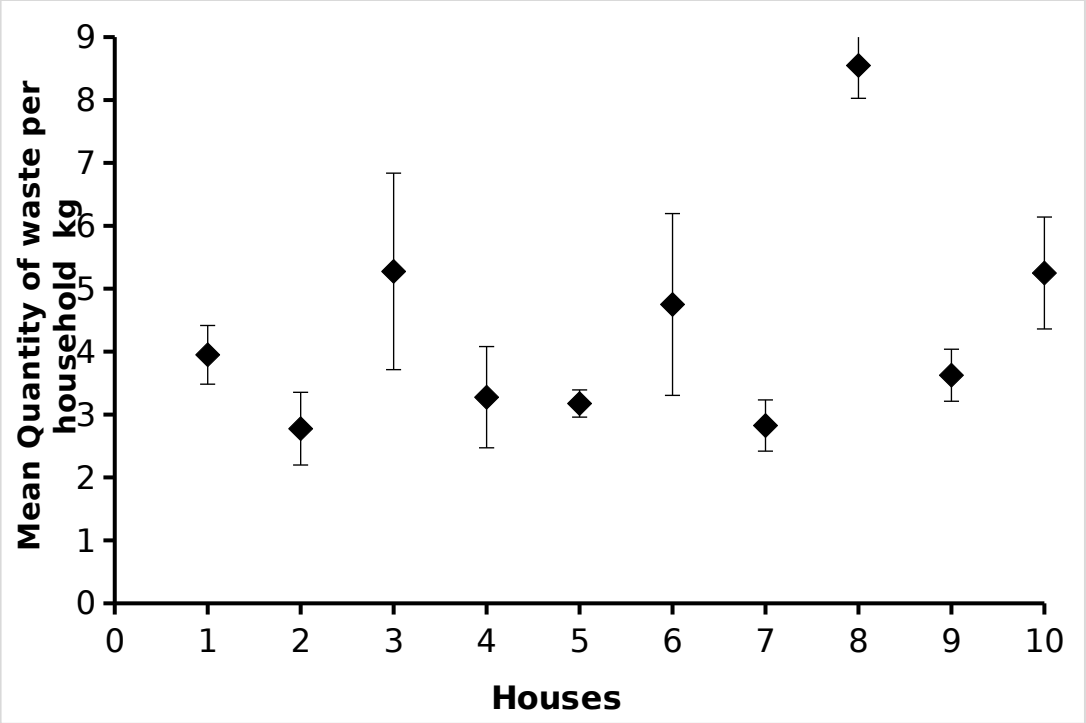

Figure 2: Quantity of Solid Waste generated in 10 selected households in the staff quarters (Gambia-Ama)

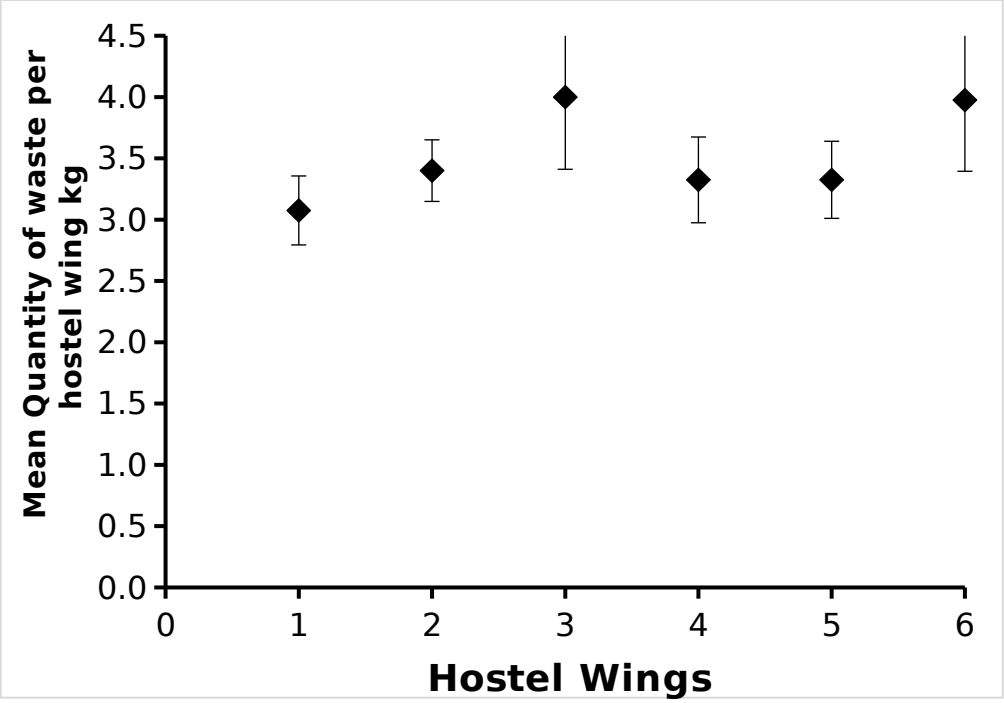

Figure 3: Quantity of Solid Waste generated in 6 wings of Block D, female students hostel at Delta Park

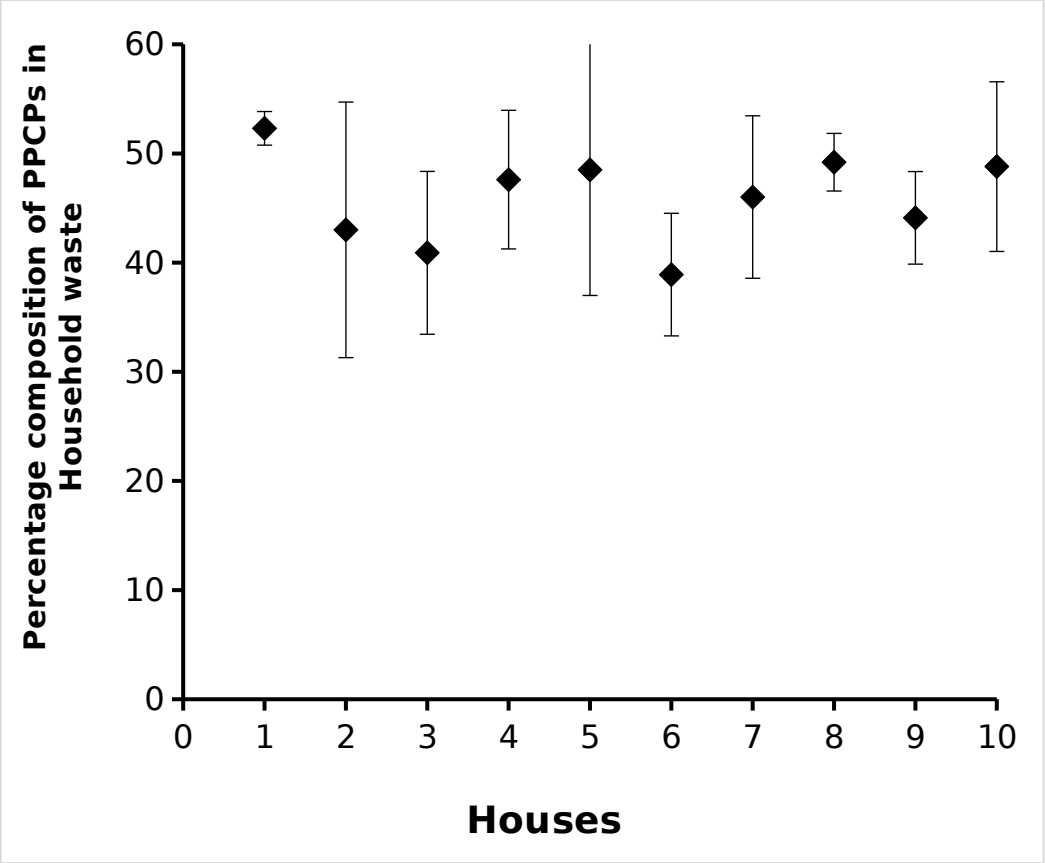

Figure 4: Relative amounts of Pharmaceuticals and Personal Care Products produced in Staff Quarters (Gambia-ama)

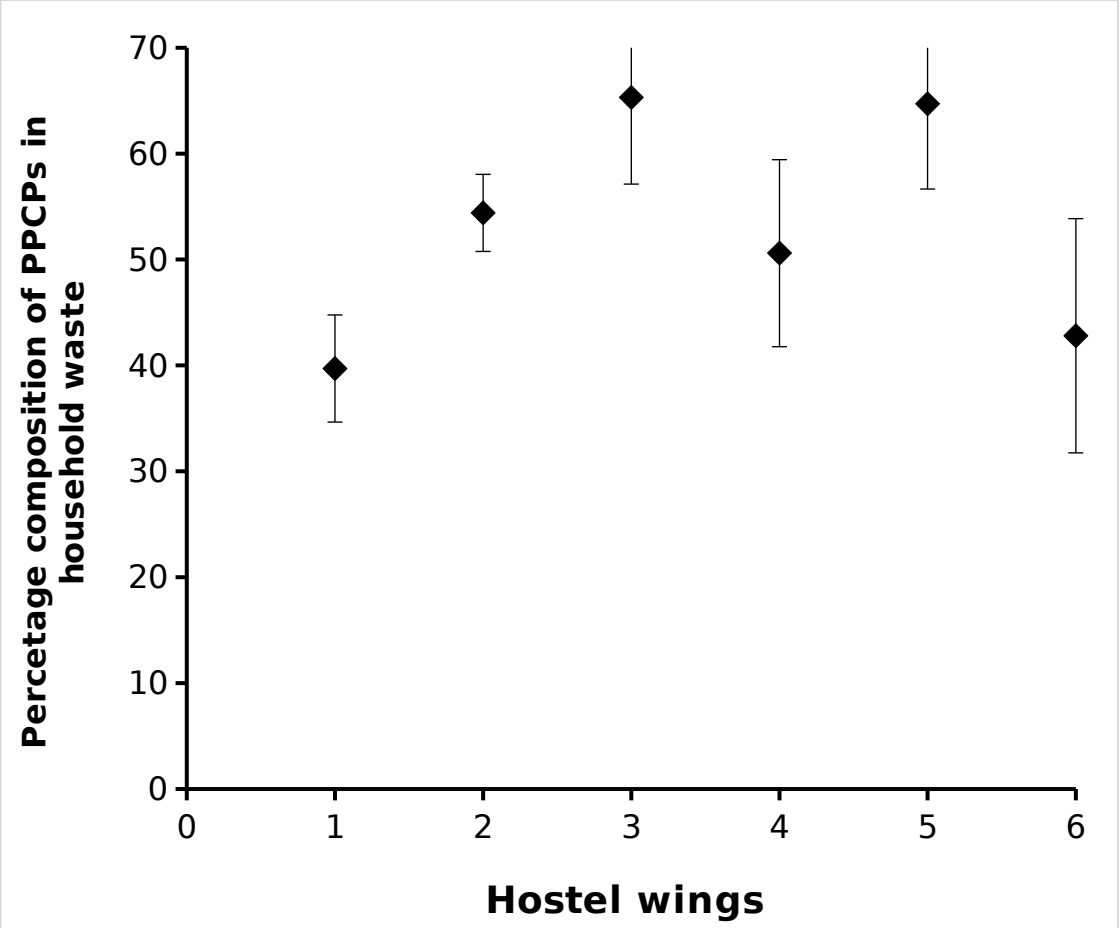

Figure 5: Relative amounts of Pharmaceuticals and Personal Care Products in students hostel (Delta Park, Block D)

Discussion

The present study showed that PPCPs were disposed off in household wastes. In the students hostel, wing C had the highest mean percentage value of PPCPs in solid waste generated (65.29%) and the least in wing A (39.69%). This simply means that the students in wing C used more of these products either as cosmetics or for health purposes than those in wing A. In the staff quarters, house 1 generated the highest amount with a percentage value of 52.26% while house 6 generated the least amount with 38.94% and this means that house 1 consumed more of these products than house 6. The results showed that the student's hostel had a higher mean percentage of 52.92% compared to the staff quarters with a mean percentage value of 45.93%. This can be explained using the age distribution between both locations. It was observed that the students hostel (female) has more young people and all females than the staff quarters which is comprised mainly of the elderly ones and a combination of both sex. Females have a higher consumption rates of these products especially as beauty care products (cosmetics and perfumes amongst others) and also single students who have the higher risk of unwanted pregnancy and infections thus will have more drugs consumption than the staff quarters which will have less use for cosmetics, pills for infections. It has been reported that between 50% to 90% of some drugs ingested is excreted as its active compound or as a conjugate metabolite (Jonathan and Nikolaos, 2005) into the toilets or directly into water ways as practice in most poor Nations like Nigeria. The disposal of drugs alongside household waste would therefore mean the active ingredients of these PPCPs are sent directly into the environment with its attendant consequences.

The occurrence of PPCPs in the environment can result to chemical interaction with other contaminants forming synergistic or antagonistic effects leading to increase chemical concentrations or emergence of more potent metabolites that may be more harmful to the environment than the parent compounds and care must be taken as even at very low levels, these chemicals have been reported to cause irreversible changes at the community level in the environment. Observed environmental effects include feminizing activity of endocrine-disrupting compounds such as oral contraceptive drug 17 alpha-ethinylestradiol on fish near waste water treatment works outfall (Jobling et al., 1998). Other observations include the development of antibiotic resistant pathogens near samples susceptible to effluent containing Pharmaceutically Active Compounds (PhACs) (Schwartz et al., 2002). Over the counter OTC Antibiotics are used extensively in fish farming and animal husbandry. Rhodes et al., (2000) and Guardabassi et al., (1999) reported that bacteria recovered from the gills and intestines of the wild commercial fish captured near fish farming activities have shown high frequencies of multiple antibiotic resistance. Bacteria recovered from samples downstream of influents of industrial waste, hospital waste have been reported to show statistically significant patterns of resistance. Other laboratory studies have reported acute toxic and genotoxic effects at significantly low LC_{50} or EC_{50} in a number of test species for example, Isidori et al., (2004) reported the ecotoxicity of six antibiotics (Erythromycin, Oxytetracycline, Sulfamethoxazole, Ofloxacin, Lincomycin and Clarithromycin) on aquatic organisms.

Despite advice on pharmaceutical packaging that recommends the return of unused medicines to pharmacies, or occasionally to flush them down the toilet, the predominant method of disposal has been reported to be via household waste even in the developed countries like the United Kingdom

(Jonathan and Nikolaos, 2005) and United States of America (Daughton and Ternes 1999). The results obtained from the analysis of the paracetamol residue in household waste samples in both sample sites were 0.6702 mg/kg of waste for the students hostel and 0.6569 mg/kg for the staff quarters. These values are statistically significant at $P<0.05$ when compared with the values in the standard drug granules. This means that the concentration of paracetamol residues found in the household waste generated in both study sites were higher than those found in the pure culture. Thus, the drug could easily find its way into the environment when the household waste is finally disposed off in dumpsites and leach, contaminating surface and ground water as well as interfere with microbial communities in soils. A risk assessment and management process of household disposal of medicine is worthy of consideration to avoid environmental contaminations by drugs and other pharmaceutical and personal care products.

Discarded pharmaceuticals are defined in the United Kingdom by the Controlled Waste Regulations 1992 [Her Majesty's Stationery Office (HMSO) 1992] as clinical waste and as such are controlled by the Special Waste Regulations 1996 (HMSO 1996). According to this legislation, such waste may be disposed of in landfill sites designed to accommodate hazardous waste, or it may be incinerated. However, once dispensed to or purchased by a member of the public, any unwanted pharmaceutical products are classified as household waste, and their disposal is not subject to any controls. Manufacturer packaging usually recommends disposal by returning to the pharmacist; however, disposal via the sink/toilet or in normal household waste is common. Pharmaceuticals in landfill sites are subject to biological degradation processes, but some may persist and even leach into surrounding groundwater and rivers (Ahel et al. 1998; Holm et al. 1995; Schwarzbauer et al. 2002).

Results obtained from the present study agrees with reports from other places such as the investigation into the disposal habits of the American public found that only 1.4% of the people they surveyed returned unused medication to the pharmacy, whereas 54% threw them away and 35.4% disposed of them in the sink/toilet (Kuspis and Krenzelok 1996). In United Kingdom, Jonathan and Nikolaos (2005) reported that two-thirds of medications (63.2%) were disposed off in household waste and household waste constitute an important pathway for the exit of PPCPs into the environment.

Conclusion

The present study showed clearly that household waste is an important pathway for disposal of PPCPs into the environment and indeed the active ingredient of these chemicals as shown by the paracetamol residue enter the environment through this pathway. The occurrence of PPCPs in the environment even at low levels has been reported to cause irreversible changes even at the genetic level. An alternative route of disposal of medicine and personal care products is advocated which may include return of medicine to producers for appropriate disposal in an integrated green pharmacy approach.

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