Pro Gradu - Master's Thesis

Occurrence of selected pharmaceuticals in groundwater, surface water, wastewater and source-separated urine in a peri-urban area of Lusaka, Zambia

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ABSTRACT

The occurrence of selected pharmaceuticals in the hydrological cycle of a peri-urban area and in urine determined for fertilizer use was studied in Lusaka, Zambia. The objective of this thesis was to determine the concentrations of antibiotics and antiretroviral drugs in groundwater, wastewater, surface water and in source-separated urine. The study area is described by insufficient water and sanitation infrastructure. Antibiotics and antiretrovirals were chosen as the target pharmaceuticals because of their vast usage for treatment of HIV/AIDS and other diseases in Lusaka and because of the potential risks they pose when entering the environment, such as ecotoxicological impacts and antimicrobial resistance.

The occurrence of the selected pharmaceuticals was investigated by collecting 26 groundwater samples, 5 surface and wastewater samples and 10 samples of source-separated urine. The samples were prepared with solid phase extraction and analyzed with liquid-chromatography-tandem-mass-spectrometry (LC-MS/MS). The studied antibiotics were amoxicillin (AMO), doxycycline (DOX), norfloxacin (NOR), ciprofloxacin (CIP), sulfamethoxazole (SMX), tetracycline (TET) and trimethoprim (TMP), whereas the studied antiretroviral drugs were lamivudine (3TC), nevirapine (NVP) and zidovudine (ZDV). Data was also collected by background information surveys and observation.

The studied pharmaceuticals were detected in the groundwater samples in relatively low concentrations with a maximum concentration of 880 ng/L of AMO. In surface and wastewater samples all studied pharmaceuticals were detected in the ng or µg range with SMX detected in surface water at a concentration of 11800 ng/L and in wastewater at 33300 ng/L, whereas the antiretroviral 3TC was detected at 49700 ng/L and 232920 ng/L. The pharmaceuticals were detected from source-separated urine in µg and mg range with TMP, 3TC and SMX found in the highest concentrations: 12800 µg/L, 10010 µg/L and 7740 µg/L. The results suggest that the high environmental occurrence of pharmaceuticals in Lusaka can be attributed to insufficient sanitation infrastructure and vast usage of medicines consequential to high disease prevalence. The source-separated urine cannot be recommended to be used as a fertilizer, but the collection of the urine would enable safer sanitation and either its appropriate disposal or treatment into fertilizer products.

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TIIVISTELMÄ

Tässä työssä tutkittiin valittujen lääkeaineiden esiintymistä kaupungin reuna-alueiden hydrologisessa kierrossa sekä lannoitekäyttöön tarkoitetussa ihmisvirtsassa Lusakassa, Sambiassa. Työn tarkoituksena oli selvittää antibioottien ja antiretroviraalien pitoisuuksia alueen pohjavedessä, jätevesissä ja vastaanottavissa pintavesistöissä sekä kuivakäymälöissä erotellussa virtsassa. Päämääränä oli myös esittää suosituksia virtsan lannoitekäytön järjestämiseksi ja ympäristöriskien vähentämiseksi.

Tutkimusalueena oli Lusakan reuna-alueella sijaitseva epävirallinen asuinalue Madimba ja sen lähiympäristö. Alueen vesi- ja sanitaatioinfrastruktuuri on puutteellista: viemäriverkostoa ei ole, ja vaikka vesijohtoverkosto onkin viime vuosina osittain laajentunut alueelle, ottavat useimmat asukkaat yhä talousvetensä pihoille kaivetuista matalista pohjavesikaivoista. Osa käyttää pohjavettä myös juomavetenä. Kuoppakäymälät ovat yleisin sanitaatiomuoto, mikä aiheuttaa käymälöiden läheisyyteen kaivettujen kaivojen saastumista etenkin sadekaudella. Pohjavesi on suuressa osassa Madimbaa hyvin lähellä maanpintaa, mikä tarjoaa määrällisesti riittävästi vettä, mutta vaarantaa veden laadun saastumisen seurauksena. Jätevedet lasketaan usein suoraan ympäristöön tai johdetaan vesistöihin ja niitä käytetään myös viljelysten kasteluun.

Madimbaan on kehitysyhteistyöhankkeiden aikana rakennettu erottelevia kuivakäymälöitä, joista saatua virtsaa ja kiinteää käymäläjätettä on ollut tarkoitus käyttää erikseen lannoitteena alueen puutarhoissa. Erityisesti virtsan lannoitekäyttö olisi ravinteiden kierrätyksen kannalta järkevää, sillä virtsa sisältää sopivassa suhteessa kasvien tarvitsemia ravinteita ja sen hygienisointi on yksinkertaista. Virtsaan erittyvät lääkeainejäämät saattavat kuitenkin ympäristöön päästessään altistaa terveys- ja ympäristöriskeille, sillä niiden on havaittu hajoavan hitaasti, kertyvän kasveihin ja aiheuttavan ekotoksikologisia vaikutuksia. Käymäläerotellusta virtsasta on kuitenkin mahdollista valmistaa turvallisia lannoitetuotteita käsittelemällä sitä eri menetelmillä.

Antibiootit ja antiretroviraalit valittiin tutkittaviksi lääkeaineiksi, sillä niitä käytetään Lusakassa laajalti HIV/AIDS:n sekä muiden tautien hoitoon. Joka viides lusakalainen on HIV-positiivinen ja noin 65% heistä käyttää päivittäin HIV-lääkitystä. Nautitut lääkeaineet

erittyvät suurelta osin virtsaan ja päätyvät sitä kautta ympäristöön joko suoraan kuoppakäymälöistä tai jätevesien mukana. Tämä on ympäristön sekä ympäristöterveyden kannalta huolestuttavaa, sillä erityisesti antibioottien on pieninäkin pitoisuuksina havaittu aiheuttavan paitsi ekotoksikologisia vaikutuksia erityisesti akvaattisille eliöille, myös mahdollista mikrobilääkeresistenssin kehittymistä. Lääkeaineiden tehon häviäminen pakottaisi vaihtamaan kalliimpiin lääkkeisiin vaikuttaen näin myös HIV:n kanssa elävien taloustilanteeseen, mikä olisi erityisen ongelmallista siksi, että nämä ihmiset ovat usein jo valmiiksi haavoittuvaisessa asemassa. Lääkeaineiden esiintymistä ympäristössä on tutkittu teollisuusmaissa paljon, mutta kehittyvissä maissa vielä suhteellisen vähän.

Lääkeaineiden esiintymistä tutkittiin keräämällä 26 pohjavesinäytettä tutkimusalueen pohjavesikaivoista, 5 näytettä jäte- ja pintavesistä sekä 10 näytettä tutkimusalueen julkisissa kuivakäymälöissä erotellusta virtsasta. Näytteet kerättiin kesä-heinäkuussa 2016 ja esikäsiteltiin kiinteäfaasiuutolla. Tiivistetyt ja puhdistetut näytteet kuljetettiin Jyväskylään, missä ne analysoitiin nestekromatografia-sähkösumutusionisaatio-tandemmassaspektrometrialla (SPE–LC–ESI–MS/MS). Tutkittavat antibiootit olivat amoksisilliini (AMO), doksisykliini (DOX), norfloksasiini (NOR), siprofloksasiini (CIP), sulfametoksatsoli (SMX), tetrasykliini (TET) ja trimetopriimi (TMP), kun taas tutkittavat antiretroviraalit olivat lamivudiini (3TC), nevirapiini (NVP) ja zidovudiini (ZDV). Aineistoa kerättiin myös taustatietolomakkeilla, havainnoinnilla sekä haastattelulla.

Pohjavesinäytteissä havaittiin puolet tutkituista lääkeaineista suhteellisen matalina pitoisuuksina. Maksimipitoisuutena havaittiin amoksisilliinia 880 ng litrassa. Sulfametoksatsolia havaittiin yli 42 prosentissa ja nevirapiinia yli 38 prosentissa näytteistä. Pinta- ja jätevesissä pitoisuudet taas olivat huomattavasti korkeampia kuin pohjavedessä: kaikkia lääkeaineita löytyi näytteistä ng- tai µg-luokan pitoisuuksina litrassa. Antibiooteista sulfametoksatsolia mitattiin pintavedestä 11800 ng/L ja jätevedestä 33300 ng/L. Antiretroviraaleista lamivudiinia havaittiin 49700 ng/L pintavedessä ja 232920 ng/L jätevedessä. Useimpien lääkeaineiden pinta- ja jätevesistä mitatut pitoisuudet olivat korkeampia kuin aikaisemmissa tutkimuksissa raportoidut pitoisuudet ja 100-1000 kertaa korkeampia kuin teollisuusmaissa. Erotellusta virtsasta mitattiin pinta- ja jätevesiin verrattuna moninkertaisesti korkeampia pitoisuuksia tutkittuja lääkeaineita µg- ja mg-luokan pitoisuuksina litrassa. Trimetopriimiä, lamivudiinia ja sulfametoksatsolia löydettiin korkeimpina pitoisuuksina, jotka olivat 12800 µg/L, 10010 µg/L ja 7740 µg/L. Trimetopriimiä, doksisykliiniä ja lamivudiinia löytyi kaikista tutkituista virtsanäytteistä vaihtelevina pitoisuuksina, ja siprofloksasiinia lisäksi yhdeksästä näytteestä kymmenestä.

Tulokset viittaavat siihen, että puutteellinen sanitaatioinfrastruktuuri, tiheä asutus ja tartuntatautien yleisyydestä johtuva runsas lääkkeidenkäyttö vaikuttavat lääkeaineiden korkeaan esiintymiseen Lusakan ympäristössä. Vastaavia tuloksia on saatu myös muualta Saharan eteläpuoleisesta Afrikasta. Tutkimusalueella käymäläerotellun virtsan säiliö- ja varastointikapasiteetti eivät tue suositeltua 6 kuukauden varastointiaikaa eikä virtsan runsas tuotanto ole tasapainossa kaupunkiviljelyn ravinnetarpeen kanssa. Korkeiden lääkainepitoisuuksien vuoksi käymäläeroteltua virtsaa ei voida suositella käytettäväksi sellaisenaan lannoitteena, mutta virtsan erottelu ja kerääminen mahdollistaa sen turvallisen loppusijoituksen tai käsittelyn lannoitetuotteeksi, josta lääkeainejäämät on puhdistettu ja jonka avulla on mahdollista kehittää paikallisia elinkeinoja. Tärkein keino lääkeaineiden ympäristöön, talousveteen ja ruoantuotantoketjuun päästessään aiheuttamien riskien ehkäisemiseksi on kuitenkin kattavan ja toimivan sanitaatiojärjestelmän rakentaminen ja ylläpito. Erottelevat kuivakäymälät mahdollistavat myös kestävän kehityksen periaatteiden mukaisen ravinteiden kierrätyksen, mikäli syklin turvallisuudesta huolehditaan.

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ABBREVIATIONS

3TC	Lamivudine
AIDS	Acquired Immune Deficiency Syndrome
AMO	Amoxicillin
API	Active Pharmaceutical Ingredient
ARG	Antibiotic Resistance Gene
ART	Antiretroviral Treatment
CIP	Ciprofloxacin
DOX	Doxycycline
EOC	Emerging Organic Contaminant
GDP	Gross Domestic Product
GDTF	Global Dry Toilet Association of Finland
HAART	Highly Active Antiretroviral Therapy
HDI	Human Development Index
HIV	Human Immunodeficiency Virus
K	Potassium
LC–MS/MS	Liquid Chromatography-Tandem Mass Spectrometry
LWSC	Lusaka Water and Sewerage Company
N	Nitrogen
NECOS	Network for Environmental Concerns and Solutions
NOR	Norfloxacin
NVP	Nevirapine
P	Phosphorus
PPCPs	Pharmaceuticals and Personal Care Products
S	Sulphur
SDGs	Sustainable Development Goals
SMX	Sulfamethoxazole
SPE	Solid Phase Extraction
TET	Tetracycline
TMP	Trimethoprim
UDDT	Urine-Diverting Dry Toilet
VIP	Ventilated Improved Pit Latrine
WWTP	Wastewater Treatment Plant
ZDV	Zidovudine

1 INTRODUCTION

“We forget that the water cycle
and the life cycle
are one.”
- Jacques-Yves Cousteau

Pharmaceuticals and personal care products are some of the most widely reported environmental contaminants belonging to the higher class of Emerging Organic Contaminants (EOCs). This is a relatively new group of unregulated environmental contaminants whose environmental fate and effects are still insufficiently understood. The presence of pharmaceutical traces in the environment has recently become an increasingly studied topic in the scientific community, especially because of the potential ecotoxicological risks caused by the exposure of sensitive species to these biologically active substances and the potential of causing antimicrobial resistance (Segura et al. 2015; Archundia et al. 2017). This study concerns antibiotics and antiretroviral drugs, pharmaceuticals that are used abundantly in the study area in order to treat Human Immunodeficiency Virus (HIV) and other common infectious diseases. Antibiotics are globally recognized as one of the major groups of EOCs, and antiretroviral drug residues appear to be of similar importance in sub-Saharan Africa, where this study was executed (Archundia et al. 2017; K’oreje et al. 2016).

The pharmaceuticals are continuously released into the aquatic environment through direct discharges of domestic wastewater and wastewater treatment plant effluents, either treated or untreated, and because of this constant release, even the biodegradable pharmaceuticals are pseudo-persistent in the surface waters (Archundia et al. 2017). These contaminants enter the wastewater mainly via human urine, which can contain a range of pollutants besides pharmaceutical compounds, for instance hormones and pathogens (Pronk & Koné 2009). In settings with inexistent sewage infrastructure where on-site sanitation is the main type of sanitation infrastructure, such as in the majority of the peri-urban areas in sub-Saharan Africa, pharmaceuticals and other pollutants can enter the surface and groundwater also through leakage from pit latrines and septic tanks (Grönwall et al. 2010). An increasing number of studies have pointed out the ubiquitous presence of pharmaceutical residues in not only surface and groundwater in all parts of the world but also in soil compartments, the highest concentrations often found in the developing countries of the global

South, where disease prevalence is often high, access to medication less regulated and sanitation infrastructure permits the release of wastewaters and urine into the environment (for example Segura et al. 2015; Kim et al. 2011; Archundia et al. 2017; K'oreje et al. 2016; Ngumba et al. 2016).

A safe environment is a core part of sustainable development, a base on which other development targets can be built upon. This idea is strongly present in the United Nations Sustainable Development Goals (SDGs) and in the Agenda 2030 (UN General Assembly 2015). At least seven out of the 17 SDGs are directly related to this study: Clean Water and Sanitation, Life Below Water, Life On Land, Good Health and Well-Being, Sustainable Cities and Communities, Reduced Inequalities and Responsible Consumption and Production. Pharmaceutical concentrations in the environment can pose toxic effects on both aquatic and terrestrial life, but they also affect the sustainability of the communities living in such environment by exposing the residents to health hazards and socio-economical risks for instance through development of antimicrobial-resistant pathogens (WHO 2017a). In addition, the populations living in locations with high pharmaceutical contamination levels are often the already vulnerable poor urban and peri-urban residents of low and lower-middle income countries in the global South (Segura et al. 2015), which can be seen as global inequality. It is becoming evident that pharmaceutical contamination in the environment is an important piece in the puzzle of sustainable development.

1.1 Motivation and need for the study

There is a large gap in the knowledge on the occurrence of pharmaceutical contamination in Africa and other developing countries, as the topic has been studied mostly in the industrial countries of the global North. A few recent studies exist covering locations in Kenya, South Africa, Mozambique and Ghana (K'oreje et al. 2016; Ngumba et al. 2016; Segura et al. 2015), but to the current knowledge of the author, the subject has not been previously studied in Zambia despite other studies concerning water pollutants in the country (for example Sorensen et al. 2015; Nkhuwa et al. 2008). This work provides unique knowledge on the occurrence of pharmaceutical contamination in groundwater, surface water, wastewater and source-separated urine in a peri-urban setting in Zambia. The intertwined issue has been recognized as an ever more serious challenge to the environment and human health, with possible ramifications to the socio-economic situations of the urban and peri-urban populations in the global South. Moreover, the antiretroviral drug concentrations in the environment are faintly studied even in the global scale, and this study adds to the scarce existing knowledge on these pharmaceuticals.

In addition, the studied source-separated urine is used as an agricultural fertilizer in the study area, and has been promoted during several development cooperation projects funded by the Ministry for Foreign Affairs of Finland and implemented by non-governmental organizations (GDTF 2017a; 2017b.). Despite this encouragement, the environmental-chemical safety of this measure has not been extensively examined as the concerns are relatively recent. Also little considerable research on agricultural measures in peri-urban areas exists, and emerging environmental issues in the peri-urban areas have been studied even less (McGregor et al. 2006). Urine is often promoted as a sustainable, low-risk and high-nutrient fertilizer in the global South (Andersson 2015), but there is insufficient knowledge on the viability of its usage in areas with high disease prevalence, including HIV, and corresponding levels of pharmaceutical consumption leading to elevated levels of pharmaceutical traces in the urine. This work produced new data on the situation in the peri-urban area of Lusaka, Zambia, and the produced knowledge can be used for assessment of agricultural use of source-separated urine in Lusaka and in similar settings with high prevalence of HIV and other diseases that require regular and voluminous use of medicines.

1.2 Research objectives

In this thesis, the occurrence of pharmaceutical concentrations in groundwater, surface water and wastewater as well as in source-separated urine in the peri-urban settlement of Madimba in Lusaka, Zambia is studied. The objective is to produce knowledge about the pharmaceutical concentrations in the environment and source-separated urine in order to enable future considerations and assessment of the possible risks they present regarding the existing sanitation systems and safe agricultural use of urine in the study area and in other similar settings. The research questions are as follows:

1. What are the measured concentrations of the selected antibiotics and antiretroviral drugs in groundwater, surface water and wastewater in the study area?

2. What are the concentrations of the selected antibiotics and antiretroviral drugs in source-separated urine in the study area?

This study also aims at presenting viable options for control and management of the pharmaceutical contamination in the environment in the study area. This is done by exploring options for safe sanitation systems, including collection and treatment of human urine, which is the main source of pharmaceutical contamination in the environment in the study area and in similar settings.

2 BACKGROUND

2.1 Lusaka, Zambia as a research context

Zambia is a landlocked country located in sub-Saharan Africa, lying on the southern African central plateau and covering a land area of 752 612 square kilometers (Zambia Country Analysis 2015). The capital of Zambia is Lusaka, whose location is presented in the map of Figure 1. together with the location of the country. Zambia has many neighboring countries as it borders the Democratic Republic of Congo to the north, Tanzania to the northeast, Malawi to the east, Mozambique to the southeast, Zimbabwe and Botswana to the south, Namibia to the southwest, and Angola to the west. The country is divided into 10 provinces, one of them being the predominantly urban Lusaka Province. With 40 percent of the population living in urban areas, Zambia is one of the most urbanized nations in sub-Saharan Africa (CSO 2014).

Figure 1. Zambia and its capital Lusaka are located in southern Africa (Wikimedia Commons 2017).

Zambia features a tropical climate with three distinct seasons: a cool dry winter season from May to August, a hot dry summer season from August to October, and a warm wet season from November to April. Rainfall is more abundant in the Northern parts of the country with an average rainfall of 1100 mm to 1400 mm per year, while the Southern and Eastern parts of Zambia are drier with 600 mm to 1100 mm of annual rainfall with occasional droughts (CSO 2014). Due to the location of Lusaka 1260 m above sea level, the capital region has a sub-tropical climate and an average annual rainfall of 865 mm (Mpamba et al. 2008a).

The population of Zambia is rapidly growing at a rate of 3.0% per year (CSO 2017), and has been estimated to have reached 17 million (IMF 2017). The life expectancy at birth for Zambian people is merely 53.3 years (CSO 2017), which, according to Karlsson & Pichler (2015), can be at least partly due to the HIV/AIDS pandemic as the evidence from some sub-Saharan countries neighboring Zambia with high HIV prevalence rates shows. 54.4 percent of the Zambian population is living below the national poverty line, most of them (40.8% of the population) living in extreme poverty. In urban areas the poverty levels are lower with 23.4% of the population living below the poverty line, compared to 76.6% in rural areas. The population of Zambia consists of 73 Bantu-speaking ethnic groups with seven major languages: Bemba, Kaonde, Lozi, Lunda, Luvale, Nyanja and Tonga. English is the official language of the country. The population is young, with 65% of Zambians below 25 years old (CSO 2016).

The Human Development Index (HDI) of Zambia was 0.579 in 2016 (UNDP 2016a). This ranking places Zambia in the medium human development category, even though the country remains in the bottom quartile of the global HDI rankings (UNDP 2016b): Zambia shares the 139th place out of 188 economies together with Bangladesh and Ghana (UNDP 2016a). The state of human development is not equally divided, as only four provinces consisting of economically active urban areas would be classified in the medium human development category, while the rural provinces would be areas of low human development (UNDP 2016b).

The Zambian economy has been heavily relying on the copper mining industry since the country’s independence from Britain in 1964. A sharp decline in the copper prices deteriorated the economy and increased poverty among Zambians from the mid 1970’s onwards, but since the mid 1990’s, the economy has seen positive growth and improving living standards after economic recovery programmes and social and economic development plans were implemented. Currently the mining sector accounts for 8% of the Zambian Gross Domestic Product (GDP) together with manufacturing, construction being the largest contributing sector with 16% of the GDP. Also rural agriculture contributes to the GDP with a 9% share (CSO 2014).

Copper is the main export product accounting for approximately 80% of the total exports of Zambia (WTO 2017), but the industry employs less than 2 percent of the population (Zambia Country Analysis 2015). The GDP per capita is 1342 US dollars (IMF 2017) and the GDP is growing at a rate of 4.2% annually (2017 projection). This consistent and robust economic growth has made Zambia a lower middle-income country, yet the growing prosperity has not managed to lift large parts of the population from poverty (Zambia Country Analysis 2015). Low copper prices and debilitating electricity supply deficits have recently been affecting Zambia's economy negatively (AfDB 2017).

Lusaka, the bustling capital of Zambia, is also the largest city of the country with a population of approximately 2.3 million (CSO 2015). The city is situated in the south-central part of Zambia, at 1280 meters above sea level covering an area of 375 km^2 of mostly flat terrain. Lusaka is the economic, cultural and transportation centre of Zambia, with a diverse population consisting of people from all the Zambian ethnic groups as well as minorities of European and Asian origin and refugees from other African countries (UN-Habitat 2007; Nyamazana et al. 2017). The population growth rate in the metropolitan area is faster than the country average, estimated at 4.0% per annum (CSO 2017), resulting from rural-urban migration and internal population growth (UN-Habitat 2007). Lusaka Province has a higher HDI 0.603 compared to other parts of Zambia (UNDP 2016b), with 11% of the population living in extreme poverty, which is much lower than the country average (CSO 2016). The city of Lusaka is challenged by weak local governance and several related problems related to rapid population growth, high levels of urbanization and unemployment, insufficient services and inadequate waste management (UN-Habitat 2007).

Despite the higher living standards in the capital region, 75-78% of the inhabitants of Lusaka are living in high-density and low-income unplanned settlements often with inadequate infrastructure and services, including sanitation facilities and water supply (Nkhuwa et al. 2006; CSO 2016). The population density in Lusaka Province is 126.8 persons per square kilometer, which is higher than the country's average density of 20.6 persons per km^2. In Lusaka, a direct link between income levels and population density exists, with higher residential densities on the outskirts of the city and lower densities in the inner city (UN-Habitat 2007). 41 percent of the urban dwellers of Zambia own their housing unit, which means that most of them are renting their home (CSO 2016).

2.1.1 Water supply and sanitation in Zambia

According to the definition of the WHO/UNICEF Joint Monitoring Programme (JMP) for Water Supply, Sanitation and Hygiene (2017), improved drinking water sources are those that “by nature of their design and construction have the potential to deliver safe water.” The criteria for safely managed drinking water are three: 1) it should be accessible on premises, 2) water should be available when needed, and 3) the water supplied should be free from contamination. Basic drinking water service is achieved when drinking water from an improved source can be collected in less than 30 minutes for a round trip, including queuing. Improved sanitation facilities, on the other hand, are those “designed to hygienically separate excreta from human contact”. Within a safely managed sanitation service, the excreta can be 1) treated and disposed of in situ, 2) stored temporarily and then emptied, transported and treated off-site, or 3) transported through a sewer with wastewater and then treated off-site. An improved sanitation facility is classified as ‘basic’, if the excreta are not safely managed. These definitions in the WHO/UNICEF JMP’s ladder of safe water supply and sanitation facilities form the baseline for the monitoring of the SDGs.

Definitions of ‘improved’ drinking water sources and sanitation facilities can differ from one country to another, and national governments do not necessarily use the same definitions and data sources as the JMP – therefore several different estimates can often be found. The JMP has also updated its definition for improved drinking water source and sanitation from the previous definition used to monitor the achievement of the Millennium Development Goals, where an improved drinking water source was classified as “one that, by the nature of its construction, adequately protects the source from outside contamination, particularly faecal matter” (WHO/UNICEF JMP 2012). For instance Grönwall et al. (2010) have criticized the older definitions of improved water supply and sanitation as being flawed, since even protected groundwater wells that were previously classified as an ‘improved’ water source, can contain unsafe water: where the groundwater is contaminated or by other means not suitable for human consumption, a groundwater well previously considered as ‘improved’ offers no protection against contaminants. While the definition of an improved sanitation facility has not significantly changed, several types of latrines, including some still considered as ‘improved’, yet not those considered as ‘safely managed’, allow fecal matter to percolate out into the groundwater.

The literature is largely based on the old definitions of improved water supply and sanitation, and data generated with the new definitions is limited. The WHO/UNICEF JMP (2017) estimates that 61% of the total Zambian population had access to at least basic drinking water source in 2015. In rural areas the percentage was 44% and in urban areas 86%. The increase rate has been less than 1% per year since 2000. Of the urban population using improved water sources, 47% are using safely managed water supplies and in 89% of cases the water is estimated free from contamination. Less than a third (31%) of all Zambians has access to at least basic sanitation facilities, 19% of rural populations and 49% of urban dwellers. The sanitation coverage in the urban areas has regressed since 2000, when 51% of urban populations had access to at least basic sanitation. Of the urban populations using improved sanitation facilities (shared facilities excluded), 25% are estimated to use improved latrines, while 8% use septic tanks and 16% have a sewer connection. In rural Zambia, 25% practice open defecation while the figure is 1% in the urban areas.

The water supply and sanitation situation varies greatly between the rural and urban areas of Zambia. 65% of the total population had access to an improved water source in 2014, while over half of the rural population uses surface water and other unimproved water sources (UNDP 2016b; Zambia Country Analysis 2016). According to UNDP (2016b), only 27% of the population has access to basic sanitation facilities, while Zambia Country Analysis (2016) presents 80% of Zambians using pit latrines as toilet facilities. As a pit latrine with a slab is considered an improved and basic sanitation facility that separates excreta from human contact in a hygienic manner (Tilley et al. 2014), the majority of Zambians appears to be using open pit latrines without slabs. According to Zambia Country Analysis (2016), the proportion of population living without access to improved sanitation has increased drastically in the last decades: from 26% in 1991 to 67% in 2010. This could be explained by worsening sanitary conditions and population growth in the peri-urban settlements of Zambia’s cities (ibid).

According to NWASCO (2016), basic sanitation coverage among the urban population of Zambia is 62.6%. NWASCO also states that more than half (55.6%) of the urban sanitation was done through septic tank systems, but Zambia Country Analysis (2016) presents that 65% of the urban population uses pit latrines. In the Lusaka Province, 74% of the urban population has access to basic sanitation according to NWASCO (2016). On the other hand, Nkhuwa et al. (2006) state that 80% of the residents of Lusaka were living with inadequate sanitation facilities in 2006. Literature on the subject is highly controversial and different definitions are used by different entities. Nevertheless, Lusaka has three major types of sanitation services: waterborne sewer systems, septic tanks, and pit latrines (Picture 1.), the latter being used by 90% of the households in unplanned settlements of the city (UN-Habitat 2007). According to UN-Habitat (2007), 3% do not have access to toilet facilities and use either communal toilets or practice open defecation. Pit latrines are often shared by several households, what makes them fill up quickly. The locations of the latrines are often inaccessible for suction tank trucks, which results in the latrines not being emptied (UN-Habitat 2007).

Picture 1. Pit latrines and hand-dug groundwater wells are common water supply and sanitation facilities in Zambia.

What comes to water supply, in 2010, 36% of the urban households of Zambia had piped water services on the premises (Zambia Country Analysis 2016) while according to NWASCO (2016), six years later almost two thirds of the urban population was served with water network connections and 83.5% of the urban population had access to a safe and reliable water source. In Lusaka Province the water supply coverage was also 83% in 2016, meaning that the proportion of inhabitants without a safe and reliable water supply would have been 17% (NWASCO 2016). However, according to Nkhuwa et al. (2006), 45% of the residents of Lusaka city did not have access to a safe water supply at the time of the study. This is in line with the WHO/UNICEF JMP (2017) results and somewhat in line with the previous reports of NWASCO (2008), stating that the urban water supply coverage was 67.9% in 2006-2007. Furthermore, Grönwall et al. (2010) suggest that many more urban residents depend on hand-dug wells than the reported 17-18% in urban Zambia. This could result from shallow well users not wanting to admit using shallow wells, as the official policy is to ban the use of them (ibid.) and they are generally considered unsafe (UN-Habitat 2007).

Lusaka, together with five other districts in the region, is serviced by the Lusaka Water and Sewerage Company (LWSC) water utility, which started its operation in 1989. The company reported 97 000 water connections in 2016 and it is running several projects to improve water and sanitation services. However, the water and sewage infrastructure is in need of major rehabilitation and expansion and nearly half (49%) of the water is lost after being produced (NWASCO 2016). This results from leakages and bursting of old pipes but also illegal water connections and vandalism (UN-Habitat 2007). The LWSC also operates 480 km of sewer lines, eight pump stations and seven wastewater treatment facilities in Lusaka (Brown et al. 2012). According to Mpamba et al. (2008a), the city has historically been entirely dependent on groundwater, while today Lusaka relies on both surface and groundwater as raw water sources: half of the water of the LWSC is abstracted from the local groundwater aquifers while the other half is imported from Kafue River as treated surface water. With the thousands of both registered and unregistered borehole wells estimated in Lusaka, the city derives approximately 70% of its water requirements from groundwater sourced from the karstic Lusaka aquifer (Mpamba et al. 2008a; De Waele et al. 2013).

Groundwater is the cheapest and obvious source of water for most water users in Lusaka, as streams are rare and often non-perennial and water demand exceeds the commercial water production (Mpamba et al. 2008b). Those city residents without access to water supply services have constructed hand-dug groundwater wells (Picture 1.) where water is drawn by hand, or private boreholes. The vast use of groundwater has been setting a large pressure on the Lusaka aquifer, and the contamination levels of the vulnerable aquifer together with waterborne diseases have been increasing (Nkhuwa et al. 2008). Lusaka has an extremely shallow groundwater table and Grönwall et al. (2010) describe the groundwater “as easily polluted as surface water in a stream”. The shallow wells and the boreholes are often unprotected, which can result in severe health risks when there is contamination from pit latrines. The water from the local shallow wells and boreholes is rarely treated and is generally considered unsafe (UN-Habitat 2007).

Nkhuwa et al. (2006) state that the use of groundwater in the whole Lusaka is increasing rapidly, as the alternatives are becoming scarcer with the population growth that is concentrating especially on the groundwater-dependent low-income areas. Also rural-urban migration is adding to the population numbers and water footprint of the city. According to

UN-Habitat (2007), the uncontrolled drilling of boreholes coupled with inadequate waste management and poor sanitation are leading to growing outbreaks of epidemics due to pathogenic groundwater pollution, especially in low-income areas. In addition, the groundwater quality in Lusaka is affected by low concentrations of dissolved arsenic related to the carbonate dolomite rock type, as well as mercury and fluoride levels exceeding WHO guidelines, as limited data of British Geological Survey (2001) suggests. Cemeteries, industrial leakages and nitrate contribute to the deterioration of the water quality as well (Grönwall et al. 2010). Fecal contamination is the greatest issue, but also organic contaminants have been detected in Kabwe, Zambia, being most prevalent in the hand-dug wells of low-income residential areas (Sorensen et al. 2015).

The hydrogeology of Lusaka is characterized by a groundwater supply from the underground karstic carbonate and schist aquifers known as the Lusaka Dolomite Formation, which have been exploited on a large scale from the 1950’s onwards. Besides the LWSC, private individuals, industries and agriculturists are abstracting significant amounts of groundwater from these aquifers, especially from the karstic carbonate aquifers with small amounts withdrawn from the schist aquifers (Mpamba et al. 2008a; 2008b). Mpamba et al. (2008a) suggest that the aquifer recharge includes inflow from on-site sanitation facilities such as septic tanks and pit latrines, as well as irrigated farming activities and unaccounted water from the LWSC water supply network. Greywater from washing and bathing together with blackwater from septic tanks and sewers add to groundwater contamination via aquifer recharge, however the absence of such infrastructure is often the main problem especially in informal settlements (Grönwall et al. 2010). Outflows include abstraction, natural aquifer drainage, drainage to streams and springs and evapotranspiration.

Lusaka is located on a plateau of the mid-Tertiary peneplain of Central Africa, and is characterized by the rapid entrance of rainwater into the groundwater and drained by three distinct river basins, Chunga-Mwembeshi, Chongwe and Kafue basins (Figure 2) (Mpamba et al. 2008b). The northwestern parts of Lusaka city, including the study area, are drained first into the Chunga stream followed by Mwembeshi River, and finally into Kafue River, also itself a tributary of the Zambezi River (LCC & ECZ 2008). The city lies on top of rocky schists formed of marble, dolomitic marble and quartzite that host interconnected subterranean karst conduits, caves and solution channels that form the city’s aquifers. Three rock formations are found in the Lusaka Plateau; Lusaka dolomite, Cheta and

Chunga formations, the latter being found on the northern parts of the city where the study area is located (Mpamba et al. 2008b). Borehole drilling information suggests that the carbonate rocks forming the Lusaka aquifer are 100-120 meters deep (Nkhuwa et al. 2008; Grönwall et al. 2010). The groundwater flows in the channels generally from southeast to northwest (Mpamba et al. 2008a).

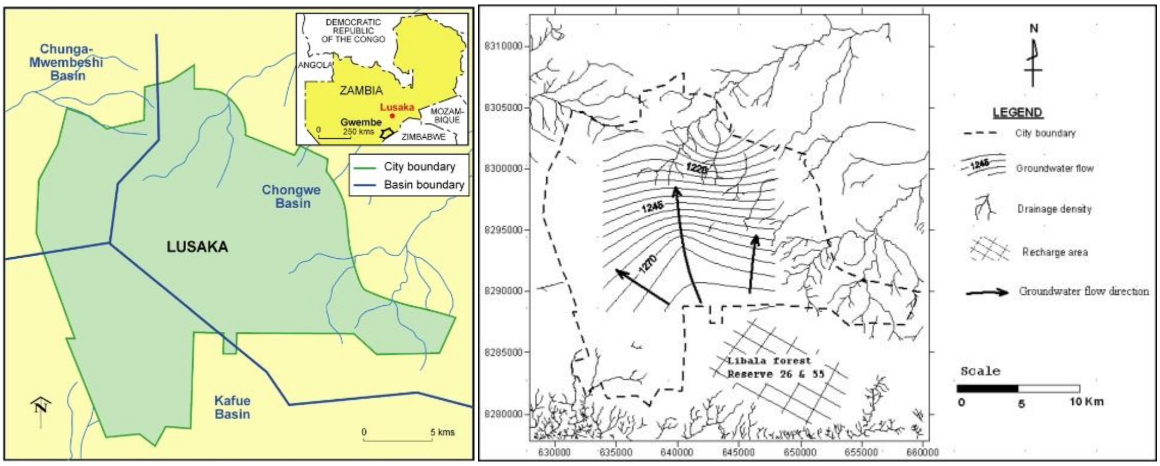


Figure 2. The urban area of Lusaka consists of three drainage basins, while the groundwater flow direction in the study area (Chunga basin) is towards west and northwest (data from the rainy season of February to April 2005) (Mpamba et al. 2008a; 2008b).

The area is described by an almost complete lack of surface drainage potential and streams, since rainwater infiltrates through the overburden of epikarst and permeable laterite or drains into the fissures and sinkholes, soon entering the groundwater (Nkhuwa et al. 2006; Mpamba et al. 2008a). The soil is coarse and of low clay content, consisting of thin layers of leptosol in the schist areas and phaeozems covering the dolomite. The flat terrain of shallow, unconfined soil causes decreased contact time for filtering the water and rapid recharge of the aquifers (Grönwall et al. 2010). According to a study conducted by Chabala et al. (2014) in Lusaka Province, the soils in the area are acidic with soil pH differing between 4.02 and 5.56. These complex hydrogeological and environmental conditions make the Lusaka aquifer extremely vulnerable to contamination and impose restrictions on land use possibilities (Nkhuwa et al. 2008; Mpamba et al. 2008a; De Waele et al. 2013). However, the unplanned city growth has resulted in industrial and housing areas developing on top of the highly vulnerable aquifers that should have been protected. Special attention should be paid to preserving the forested main recharge area located southwest of the city from anthropogenic activities (Mpamba et al. 2008a).

Contamination of groundwater aquifers is not the only issue related to water supply, as Zambia is also affected by water insecurity. Mpamba et al. (2008b) suggest that annual groundwater abstraction volumes have already exceeded the recharge, which is estimated between 8-35% of the annual rainfall. The raw water resources of Zambia are diminishing also because of climate change (NWASCO 2016). According to USAID (2007), the renewable water resources per capita have dropped from 8726 m^3/person/year in 1960-2007 to 7430 m^3/person/year in 2015 (projection). In the recent years, many private and commercial boreholes in Lusaka city have experienced “reduced yields as low as zero” according to NWASCO (2016), which has affected the water supply service hours of certain areas. This is one sign of the water insecurity affecting Zambia, caused not only by climate change, but also by poor urban planning and waste management, industrial expansion, rapid urbanization and population growth and the slow pace of water reforms.

Zambia has already had to start rationing its water supply and introduce electricity shortage load shedding schemes because of the inability to produce enough hydroelectricity with its decreasing water resources (NWASCO 2016). This has had an effect on the industry and economy of the country (IMF 2017). A more systematic monitoring, management and sharing of data on groundwater would help to manage the water resources of Zambia more sustainably and improve the water supply in the peri-urban areas of Lusaka (Mpamba et al. 2008b; Nussbaumer et al. 2016).

2.1.2 Peri-urban areas in Lusaka

The study area of this thesis research is the informal peri-urban community of Madimba in Lusaka. The growing cities of global South are often surrounded by densely habituated and impoverished shantytowns and other informal settlements, where infrastructure, service provision and shelter security are inadequate (McGregor et al. 2006). Also the peri-urban areas of the Zambian capital Lusaka are characterized by such unplanned, informal settlements, which are often called ‘compounds’ in the Zambian context. The UN-Habitat (2015) defines informal settlements as “residential areas where 1) inhabitants have no security of tenure vis-à-vis the land or dwellings they inhabit, with modalities ranging from squatting to informal rental housing, 2) the neighbourhoods usually lack, or are cut off from, basic services and city infrastructure and 3) the housing may not comply with current planning and building regulations, and is often situated in geographically and environmentally hazardous areas."

In the global South, especially in sub-Saharan Africa, the informal settlements are often situated in the peri-urban interface, a transition zone between the traditional meanings of 'urban' and 'rural' where no clear line between the two exists (McGregor et al. 2006). Defining peri-urban areas has its challenges, yet Phillips et al. (1999) have included a definition of the peri-urban interface from ODA's Renewable Natural Resources Research Strategy:

"The peri-urban interface is characterised by strong urban influences, easy access to markets, services and other inputs, ready supplies of labour, but relative shortages of land and risks from pollution and urban growth."

According to McGregor et al. (2006), the peri-urban areas come in different sizes and varying nature, but what they have in common is combining both urban and rural processes, as well as their landscapes being subject to rapid changes under the urban pressure. The peri-urban areas are often located in the periphery of cities and characterized by a mixture of formal houses, shanties, rural huts and other buildings. Eventually they become part of the built-up urban area (Phillips et al. 1999; McGregor et al. 2006). They may be rebuilt in more formal style, but often maintain their identities and even structures of traditional chieftaincy (McGregor et al. 2006). Most cities in sub-Saharan Africa, including Lusaka, have a large proportion of their population living in informal settlements (Nchito 2007).

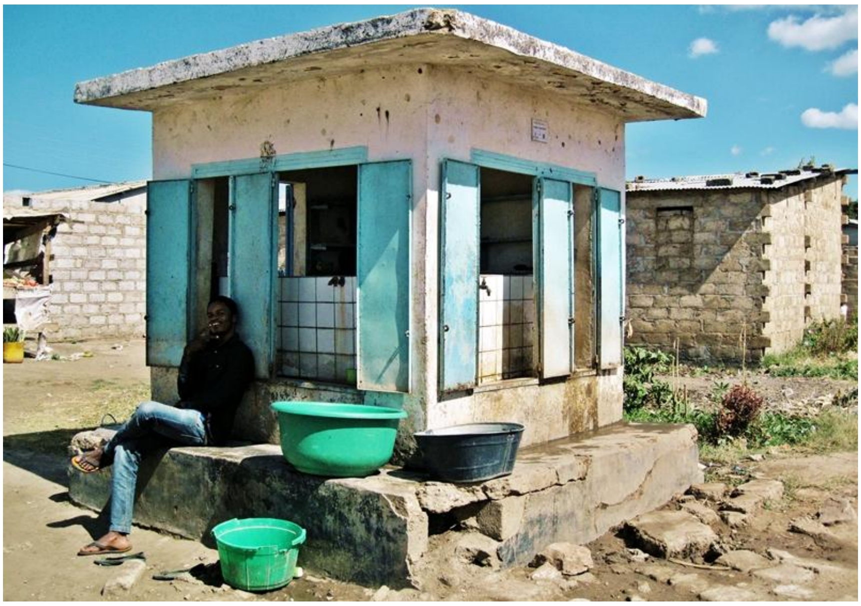
The city of Lusaka developed as a railway siding town and was first designed as a spacious, racially separated garden city for a ultimately European population of 20 000 in the early 20th century (Makasa 2010). The city planning proved ineffective with the insufficient resources of the governing body Lusaka City Council, and as a result, the areas surrounding the city center of Lusaka started growing uncontrollably without any planning guidance (Makasa 2010; UN-Habitat 2007). This unplanned expansion caused by weak local governance led to the establishment of numerous squatter settlements, which remain surrounding the city in its peri-urban areas. The settlements were first built by workers of nearby farms and industries such as quarries, and continue being the easiest and most affordable housing solution for many (Nchito 2007; Yasini 2007).

Although some sizeable unplanned settlements existed already before the independence of Zambia in 1964, they are partially a post-independence phenomenon: according to UN-Habitat (2007), the spread of such settlements are partly caused by the migration of rural people, who had been contained by colonial restriction of movement, to urban areas. The deficient housing stock caused the migrants to be unable to find affordable housing in the conventional areas and due to the inadequate land delivery systems for the poor and disadvantaged, they found their own solutions settling where they could find land (Nchito 2007). The squatters were first treated as a threat, with some attempts to demolish their houses, but gradually an official laissez-faire attitude found its way to the housing policy, stating that the areas should be upgraded instead of demolishing (Yasini 2007). Upgrades have been carried out with aid from international donors. The unplanned settlements host both low- and medium-cost housing, as some wealthier residents have built their houses according to their income (Nchito 2007).

In Lusaka, most of the unplanned settlements are situated in close proximity to the industrial areas surrounding the city center, while others are on the outskirts of the city (Yasini 2007). The settlements are generally characterized by inadequate shelter and waste management, as well as lack of services (UN-Habitat 2007). The main roads are often tarred and in good condition, while the unpaved gravel roads inside the settlements are in dreadful state, becoming impassable in the rainy season. Many, but not all, settlements have services such as schools, clinics, churches, community halls and police posts, but these are often inadequate for the high numbers of people (Yasini 2007; UN-Habitat 2007). Unemployment is high (Simwinga et al. 1997 in the World Bank 2002), but most of the residents are engaged in informal employment such as petty trading, shopkeeping, concrete block making, laundry washing, conducting, carpentry, metal and tin working, stone crushing or being a maid. Illicit occupations such as prostitution, beer brewing and theft are also somewhat common, according to Yasini (2007).

The water supplies in the informal settlements of Lusaka include tap water, boreholes and hand-dug shallow wells, the latter being used by the residents who are not able to pay for water or in the case of a water shortage (Yasini 2007; UN-Habitat 2007). In a community profiling survey carried out in nine unplanned settlements of Lusaka, less than half of the respondents said they paid for the water they use, while over 80 percent described water distribution as poor (Simwinga et al. 1997 in the World Bank 2002). Limited tap water is provided by the licensed service provider Lusaka Water and Sewerage Company or by Water Trusts, being distributed through individual, public or communal taps, the latter being paid by a certain group of users (Yasini 2007; Grönwall et al. 2010). According to UN-Habitat (2007), the LWSC provides water via the central water distribution system to the perimeter of most peri-urban settlements. Thereafter, local resident committees manage the distribution of the water. However, the tapped water supplied via this system and from local borehole systems is often inadequate and the supply is inconsistent.

According to Grönwall et al. (2010), public taps with free water used to exist in the settlements but have become very rare. Water can be acquired also from water kiosks (Picture 2.) with monthly or per container fees, hand pump wells and *dambo* (swamp) wells, as well as occasionally purchased from well owners or rarely from tanker trucks. Power cuts make the tap water supply to be erratic and available at only certain hours, thus queues can get long. Large part of the residents depend on groundwater either directly through their own shallow wells, community-based wells or vendors, or indirectly through the piped water supply, communal taps or water kiosks (Grönwall et al. 2010).



Picture 2. The peri-urban settlements of Lusaka are often serviced by water kiosks, where customers can purchase water either by monthly fee or per container.

A vast majority (90%) of the residents living in the unplanned settlements of Lusaka use unlined and unprotected pit latrines as the sanitation system and over 60% of the households share their toilet facilities (Simwinga et al. 1997 in the World Bank 2002). The pit latrines can sometimes be raised or lined, but all of them are simple constructions with a depth of 2-4 meters. They are usually covered with soil once filled (Münch & Mayumbelo 2007). According to Yasini (2007), most of the latrines are in poor condition, with a few flush toilets with septic tanks as well as some ventilated improved pit (VIP) latrines being in better shape. Some residents do not have toilets and they either use the toilets of neighbors and bars or alternatively practice open defecation into plastic bags and beer cartons (ibid).

There is no systematic waste collection in the unplanned settlements, which has lead to serious problems related to solid waste management with garbage being dumped to the roadsides, drains, pits and into the environment in general, where it attracts disease vectors, produces odors and leaches pollutants into the soil and water sources (Simwinga et al. 1997 in the World Bank 2002; Yasini 2007). The settlements are often situated in low lying areas prone to flooding, and without drainage systems they experience flash floods in the rainy season (Yasini 2007; Nchito 2007). The main roads generally have drains but these are often blocked with garbage and soil matter (Yasini 2007). The houses are predominantly made of concrete blocks with corrugated iron or asbestos sheet roofs and most of them have a backyard that fits a well. Two thirds of the community profiling survey respondents reported owning their house or plot, but only 12% had security of land tenure (Simwinga et al. 1997 in the World Bank 2002). Furthermore, the poor house structures can lead to insecure and unhealthy living environments for the residents (Yasini 2007).

Yasini (2007) describes 25 unplanned settlements in Lusaka, but according to UN-Habitat (2007), over 35 of the unplanned settlements of Lusaka had already been regularized as “Improvement Areas” under the Housing Act (Cap 194) by 2007, with infrastructure and service developments. Nchito (2007), on the other hand states that 43 unplanned settlements existed in the city at the time, while Grönwall et al. (2010) estimate 35-40 unplanned settlements being found in Lusaka. However, many newer or smaller settlements remain unrecognized without legal status and outside of studies, including Madimba, which could be thought to be a part of Chunga, a neighboring low-income ‘site and service’ planned housing scheme area (Mwansa 2016).

The unplanned settlements are growing more rapidly than the rest of the city through densification and outward expansion and even the regularized improvement areas are facing numerous challenges (UN-Habitat 2007; Nchito 2007). While shallow wells provide a water source at close proximity, the insufficient or non-existent basic infrastructure and poor environmental conditions such as the inadequate sanitation facilities, drainage and solid waste disposal along with poor hygiene awareness are among the main problems and make the residents of the unplanned urban settlements vulnerable to epidemics (UN-Habitat 2007; Grönwall et al. 2010). Also water treatment could be improved as endemic diarrheal disease and regular cholera outbreaks are yet to be tackled with widespread use of chlorine or other water treatment methods despite the subsidized prices and occasional free distribution (Grönwall et al. 2010). Provision of safe water and sustainable sanitation for the urban poor would be vital, but insecure land tenure, lack of political will, financing, cost recovery and limited choice of technical options pose challenges to these developments (Münch & Mayumbelo 2007).

2.1.3 Dry sanitation projects in Lusaka

Finland has been supporting dry sanitation projects in the Lusakan peri-urban community of Madimba since the year 2008. According to Kawanga (2005), no other donor interventions had been implemented in the community before that, besides installation of communal water taps in the community by the Japan International Cooperation Agency (JICA). The research for this thesis was conducted in collaboration with the most recent of these projects, the Innovative Sanitation for Peri Urban Areas in Lusaka project, which was implemented between the years 2014 and 2016. It was a joint development cooperation project between the Zambian non-governmental organization Network for Environmental Concerns and Solutions (NECOS) and their international partner The Global Dry Toilet Association of Finland (GDTF). The main donor of the project was the Ministry for Foreign Affairs of Finland (GDTF 2017b).

The Innovative Sanitation for Peri Urban Areas in Lusaka project was working in four constituencies and eight wards in the Lusaka metropolitan area. The combined population of the project areas was estimated at 18 000 people, one third of them living in the study area of Madimba. The project promoted equal access to clean water, better sanitation and safe environment for these communities. The project activities included construction of 150 dry toilets, but also facilitating policy dialogue workshops as well as training and capacity building for local small and medium-size enterprises. Promotion of school WASHE and community hygiene, facilitating stakeholder meetings and developing materials and dry toilet user manuals were also included in the project activities. Awareness raising and promotion of natural nutrient cycle were key elements of the project, which aimed at improving both the living standards of the people and the state of the environment, by decreasing especially the contamination of groundwater in the project area. The Urine-Diverting Dry Toilets (Figure 3. and Picture 3.) were in the center of the project (GDTF 2017b).

The UDDT functions as a waterless toilet that separates urine and feces by diverting urine into a separate jerry can container from the user interface (toilet seat) structure through piping. The feces are allowed to dehydrate in a separate vault for pathogen reduction and further used as soil conditioner, while the urine can be used as liquid fertilizer (Tilley et al. 2014). The method can have many other benefits in addition to safe and sustainable sanitation, including increased food production and income. The UDDTs found in Madimba, Lusaka are built based on the schematic structure seen in Figure 3, except for the urine tank which built as a separate compartment behind the toilet and equipped with a hatch for ease of emptying. The toilets have two seats or squatting pans and two dehydrating vaults, one of which being in use at a time. When the chamber fills up, the other vault, filled with dehydrated fecal matter that was left to stabilize for a certain period, is emptied and taken into use. Ventilation of the toilet is ensured with a screened vent pipe.

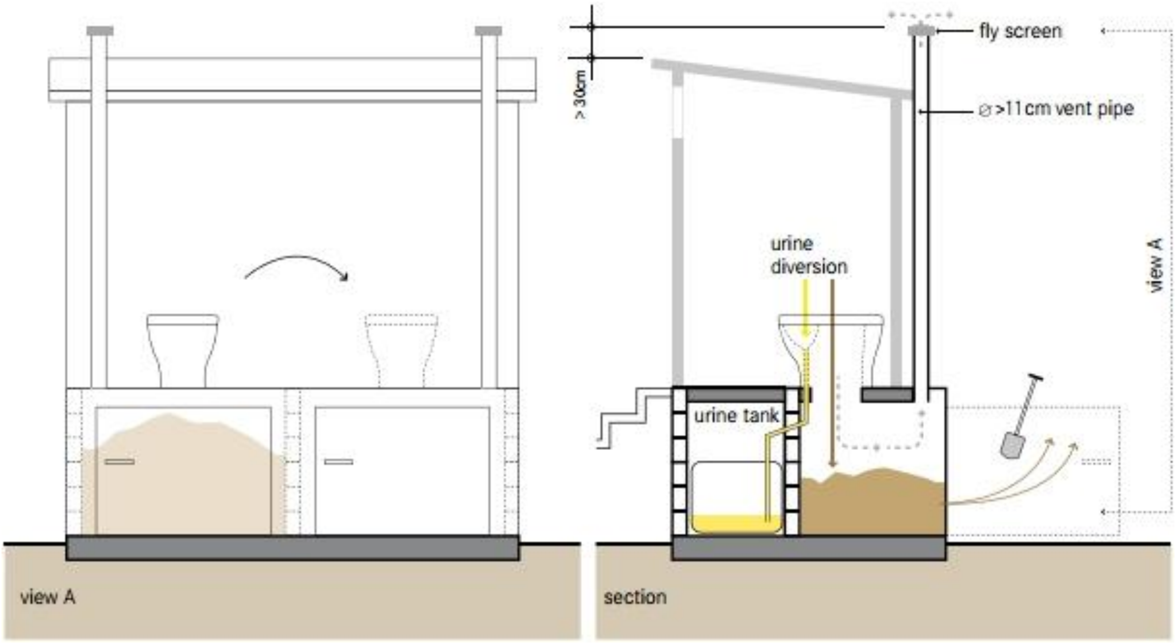


Figure 3. The schematic structure of the Urine-Diverting Dry Toilet (Tilley et al. 2014).

The Innovative Sanitation for Peri Urban Areas in Lusaka project was a continuation project to the Zambia Sustainable Sanitation Improvement Project that was initiated in 2008. This was the first sanitation development project in Madimba, which aimed at improving the sanitation of the area and increasing human wellbeing and hygiene awareness. This was achieved by building dry toilets, establishing water points and organizing hygiene education. The Finnish Ministry for Foreign Affairs was the donor of this project as well (GDTF 2017a). A water kiosk serving approximately 50-80 households per day was also established. The kiosk was not in operation at the time of the data collection for this research due to the electricity cuts affecting the water pumping capacity (NECOS 2015).

Simultaneously with the Innovative Sanitation project, NECOS implemented a research project promoting the human urine drip fertigation in greenhouse agriculture. The project aimed at strengthening the urban agriculture and backyard gardening as well as waste reuse by development of a community greenhouse and capacity-building activities (NECOS 2015). The greenhouse tomatoes cultivated during the project were fertilized with hygienized urine and ‘humanure’, dried fecal matter collected from the dry toilets of Madimba. The building of the greenhouse technology with a cultivation area of 160 m^2 was supported by the Student Union of the University of Helsinki in 2013. The project organized also training for women in urban agriculture and training in sanitation service chain (Kawanga et al. 2015).

Picture 3. Several urine-diverting dry toilets have been constructed in Madimba during the sanitation projects. On the right, a pile of dried fecal matter has been emptied from the toilet.

NECOS and GDTF have also been collaborating to bridge the gaps in the peri-urban sanitation policies in order to improve sanitation management for the alleviation of poverty, improved livelihoods and environmental upgrading in Lusaka and elsewhere in Zambia. This has been done through promotion of institutionalizing the UDDT curriculum, introduction of public-private partnerships into sanitation schemes, and development of sanitation service chains and security of tenure (NECOS 2015). The work is being continued in the ongoing country-wide project Zambia Dry Sanitation Country Program during the years 2017-2020 (GDTF 2018). The GDTF (2017b) states that in the peri-urban settlements of Zambia, “there is an urgent need for provision of water and sanitation services as basic human rights and enforcement of relevant regulations”. The water and sanitation situation of these settlements needs to be improved in order to achieve sustainable development in terms of the living environment as well as livelihood, socio-economic development and poverty alleviation (GDTF 2017b; NECOS 2015).

2.1.4 HIV and ART in Zambia

Sub-Saharan Africa is at the center of the global HIV/AIDS epidemic, with 71% of the new infections occurring in the region (UNAIDS 2009). Zambia has one of the highest Human Immunodeficiency Virus (HIV) prevalence in the world, 13.3% of the adult population aged 15-49 having been HIV positive in 2014 (National AIDS Council 2015). The population of Zambia is also one of the most seriously affected by AIDS, acquired immunodeficiency syndrome, in the world (Stringer et al. 2006). Zambian women are being infected at a younger age than men (UNAIDS 2004) and among women the HIV prevalence is also higher than among men: 15.1% compared to 11.3% (National AIDS Council 2015). 1.6% of the population becomes newly infected annually, of which 2% are women and 1.2% men (Zambia National HIV/AIDS/STI/TB Council 2009). For instance in 2009 this accounted for approximately 82 000 people (Ministry of Health 2010), but in 2016 the number had dropped to approximately 59 000 new infections per year. Overall, there are 1.2 million people living with HIV/AIDS in Zambia (UNAIDS 2017).

The HIV prevalence is higher in urban areas than in rural areas, also in the capital Lusaka, where a greater percentage of the population is living with HIV than in the rural areas. Some sources state that 19.4% of the population of Lusaka is living with HIV/AIDS (CSO 2014), while others estimate the prevalence rate at 22% of the population of the city (Stringer et al. 2006). In any case, roughly one in five inhabitants of Lusaka is HIV positive. The most common cause of death in the Lusaka Province is tuberculosis along with fever/malaria both accounting for 11.7% of the total deaths, which are common infections causing mortality among HIV patients (CSO 2016).

WHO (2017) defines Antiretroviral therapy (ART) as “treatment of people infected with human immunodeficiency virus (HIV) using anti-HIV drugs”. ART is the medical management of HIV infection with combined use of at least three antiretroviral drugs in order to reduce the likelihood of resistance development in the virus. The drugs are taken on a once-daily basis. This is the most common form of treatment and such combined use of medication is often called HAART, Highly Active Antiretroviral Therapy, as it suppresses the HIV replication (WHO 2017a). The goals of ART include reducing viral load, restoring and preserving immunological function, improvement of the quality of life of the patient, reduction of HIV-related morbidity and mortality, and reducing the risk of transmitting the infection onwards (John Hopkins University 2008; WHO 2017a).

Access to ART has in the past been limited to the affluent Zambians, who have been able to seek treatment through private medical practices. From the years 2004-2005, after an estimated 89 000 Zambians died of HIV/AIDS in 2003 alone (UNAIDS 2004), wide-scale HIV treatment programs were initiated by the Ministry of Health in the primary health care centers of Lusaka, supported by the US President’s Emergency Plan for AIDS Relief (PEPFAR), the Global Fund to Fight AIDS, Tuberculosis, and Malaria and other donors. The goal was to guarantee universal access to free-of-cost HIV care and treatment with antiretroviral drugs and laboratory tests provided (Stringer et al. 2006; Schumaker & Bond 2008). After switching from the paid subsidized treatment of 2004 to the universal free treatment in 2005, the public health services of Zambia are continuing to provide free antiretroviral drugs. Because of these initiatives, ART has become increasingly available (Samuels & Rutenberg 2011). However, cost of transportation to health clinics and gender roles making ART more available to men than women can be limiting factors to ART access, among others (Hardon et al. 2007). Globally, the HIV prevalence rates are still growing, but less people are dying of AIDS-related causes, which is largely because of ART (Kharsany & Karim 2016).

According to UNAIDS (2017), 65% of Zambian population living with HIV/AIDS are on medical antiretroviral treatment, which translates to approximately 800 000 people. The coverage is slightly higher in adults than in children (67% vs. 52%). WHO (2016) has removed limitations on eligibility for ART among people living with HIV and is recommending the treatment for all affected populations. Also Zambia is aiming to treat all its citizens living with the infection regardless of the CD4 cell count (UNAIDS 2017). While Zambia is home to 3% of the global HIV positive population, between 2010 and 2013, 4% of all the people receiving ART were living in Zambia (UNAIDS 2014). According to the National AIDS Council of Zambia (2015), in Lusaka Province, approximately 176 000 people were receiving ART in 2015. Nevertheless, Lusaka having a population of approximately 2 282 000 (2015 projection) (CSO 2015), the percentages of up to 22% of the population living with HIV and 65% of them receiving ART indicate that there can be up to 326 000 daily ART receivers in the capital of Zambia.

ART, including medication, counseling and testing, can be accessed at health centers offering antiretroviral treatment. Most Zambian medical institutions offering ART provide only clinical care. ART can be prescribed by health care providers that are legally recognized for prescription of drugs in Zambia, trained in HIV/AIDS management, have access to sustainable drug supply, possess facilities to monitor therapy, and who participate in training in use and monitoring of ART (John Hopkins University 2008). The Baseline survey for the peri-urban settlement of Madimba presents that in 2005, less than 5% of the residents of the community had undergone voluntary counseling and testing for HIV. This could have been largely due to the stigma it poses on urban populations. HIV/AIDS has also been identified as a cross-cutting issue that should be taken into account when promoting dry toilets (Kawanga 2005).

Antibiotics are described as medicines that are used worldwide to inhibit the growth of or destroy microorganisms such as bacteria (Oxford Dictionaries 2017b). When treating HIV, it is common to use antibiotics together with the antiretroviral medication to keep the often fatal opportunistic infections, resulting from reduced immunological defense, in control. Therefore antibiotics are often used together with antiretroviral drugs to treat latent tuberculosis and other opportunistic infections and are vital for reduction of mortality in HIV patients especially in developing countries (Jaatinen et al. 2016; Fadeyi et al. 2015). According to Maartens (2002), the opportunistic infections in HIV patients in sub-Saharan Africa differ notably from those encountered in the industrialized countries, as environmental exposure happens with different bacteria. Tuberculosis is the most common cause of morbidity and mortality in HIV patients in sub-Saharan Africa while other prevalent opportunistic virulent pathogens include *Streptococcus pneumoniae* and *Salmonella* spp., causing serious bacterial infections (Maartens 2002). Antibiotics that are used to treat tuberculosis, sometimes referred to as anti-tuberculotics, are prescribed in large doses of several hundreds of mg per day (Jaatinen et al. 2016). This research covers seven antibiotics: amoxicillin (AMO), ciprofloxacin (CIP), doxycycline (DOX), norfloxacin (NOR), sulfamethoxazole (SMX), tetracycline (TET) and trimethoprim (TMP).

Amoxicillin is a broad-spectrum semi-synthetic antibiotic related to penicillin, which is used against gram-positive cocci bacteria to treat infections such as ear infections, respiratory infections, skin infections and urinary tract infections (Castle 2007). Amoxicillin is one of the most commonly prescribed antibiotics around the world, with traces often documented in WWTP effluents (Toxnet 2017; Andreozzi et al. 2005).

Ciprofloxacin is a broad-spectrum antimicrobial of the fluoroquinolone class. It is used to treat a vast number of infections, for instance urinary tract infections, lower respiratory tract infections, diarrhea and typhoid fever (Drugbank 2017).

Doxycycline is a long-acting synthetic antibiotic and antiparasitic drug being one of the tetracycline derivatives and having similar antimicrobial activity as them. It is used for a variety of bacterial infections, including respiratory tract infections, cervicitis and prophylaxis of malaria, especially the chloroquine or sulfadoxine/pyrimethamine resistant malaria (*Plasmodium falciparum*) (Drugbank 2017; Toxnet 2017). It can also be used as an alternative to treating plague and tetanus (Drugbank 2017).

Norfloxacin is a synthetic broad-spectrum antibacterial belonging to fluoroquinolones (Drugbank 2017). It is effective against most gram-negative and gram-positive bacteria and used to treat infections such as urinary tract infections, cystitis, gonorrhea and prostatitis (Vallerand & Sanoski 2015; Drugbank 2017). NOR has been detected in WWTP influents and effluents globally, at least in Australia, China, Europe, USA and Mexico (Le-Minh et al. 2010).

Sulfamethoxazole is an antibiotic belonging to the class of sulfonamides. It is used for the treatment of a broad range of bacterial infections causing for instance bronchitis, prostatitis, diarrhea and skin infections (Drugbank 2017). SMX has been detected in surface waters, runoffs from arable land and in WWTP influents and effluents. The general population can be exposed to SMX via food and drinking water (Toxnet 2017). The broad spectrum of activity of SMX has been limited by the development of antimicrobial resistance (Drugbank 2017).

Tetracycline is a short-acting, broad-spectrum polyketide antibiotic. It is used to treat bacterial infections such as upper and lower respiratory infections, cholera and different fevers, including typhus fever and tick fevers (Drugbank 2017).

Trimethoprim is an antibacterial agent related to pyrimethamine. It is potentiated by sulfonamides and therefore often prescribed together with sulfamethoxazole. Trimethoprim is used to treat infections such as pyelonephritis, prostatitis, respiratory and ear infections and diarrhea. It can also be used alone as an antimalarial drug. Resistance to trimethoprim has been reported (Drugbank 2017).

Antiretroviral drugs are a class of medicines that “inhibit the activity of retroviruses such as HIV” (Oxford Dictionaries 2017c). Antiretrovirals are negligibly studied pharmaceuticals which are used for treatment of diseases caused by retroviruses such as HIV/AIDS (WHO 2016). They are often used as fixed-dose combinations of two or three antiretroviral drugs, especially in resource-limited settings such as Zambia (Kumar et al. 2006). The antiretroviral drugs studied in this research are lamivudine (3TC), zidovudine (ZDV or AZT) and nevirapine (NVP).

Lamivudine is a synthetic nucleoside analogue medicine that is used for the treatment of HIV infection and chronic hepatitis B. 3TC is generally used with patients at end-stage hepatitis B and patients who have failed interferon therapy. Resistance develops within a few years, resistance rates being 50% in 2 years and 90% in 4 years (Török et al. 2009).

Nevirapine is a non-nucleoside reverse transcriptase inhibitor that is used as a part of combination therapy with other antiretroviral drugs for treatment of HIV and AIDS. It belongs to the class of alkyldiarylamines. Nevirapine is generally only prescribed after the immune system has declined and it is always taken with at least one other antiretroviral drug such as lamivudine or zidovudine, because resistance can develop if the drug is taken alone. However, even with proper usage, nevirapine is effective for a limited time only (Drugbank 2017).

Zidovudine is a potent nucleoside reverse transcriptase inhibitor used in combination with other antiretroviral drugs to treat HIV infections. It acts as a chain-terminator of viral DNA during reverse transcription, improving symptoms of AIDS (Drugbank 2017).

Antiretroviral therapy is often comprised of at least three antiviral drugs and the antibiotic co-trimoxazole. Fixed-dose combinations of antiretroviral drugs are generally used as first-line regimens in sub-Saharan Africa and other developing regions. Two combinations, zidovudine/nevirapine/lamivudine (ZDV/NVP/3TC) and stavudine/nevirapine/lamivudine (D4T/NVP/3TC), are available as combination therapy regimens, and their advantages include convenience, reduction in prescription errors, reduced pill counts, and potential for improved adherence (Kumar et al. 2006).

Co-trimoxazole is a combination of sulfamethoxazole and trimethoprim. These are often used together, because bacterial resistance develops more slowly with the combination of the two drugs than with either trimethoprim or sulfamethoxazole alone. Co-trimoxazole is a feasible, well-tolerated and inexpensive therapy used to treat a variety of bacterial, fungal and protozoan infections. It also is an off-patent drug and therefore widely available even in resource-limited settings (WHO 2016), which has made it perhaps the most used antibiotic among the HIV infected populations globally. Life-threatening bacterial infections including respiratory infections, occurring in patients with advanced HIV, have been reduced because of it, especially in resource-poor settings such as Zambia (Seddon & Bhagani 2011).

A typical daily dosage of antiretroviral drugs for a person living with HIV is 150 mg of lamivudine (3TC) twice daily or 300 mg once daily, 250–300 mg of zidovudine (ZDV) twice daily and 200 mg of nevirapine (NVP) twice daily, except for the first 14 days, when this amount is taken once daily (WHO 2016). Often this is topped with the antibiotic co-trimoxazole with the daily dosage of 960 mg, which consists of 800 mg of sulfamethoxazole and 160 mg of trimethoprim (WHO 2014). This means that a person living with HIV can consume up to 1300 mg of antiretroviral drugs and 960 mg of antibiotics daily, according to recommendations. However, data on the actual consumption of antibiotics in the developing world is limited, because first-line antibiotics can be bought easily and without prescription from pharmacies, grocery stores, and even mobile drug vendors (Fadeyi et al. 2015).

Zambia relies almost completely on imported and donated pharmaceuticals to meet the local drug demand. The expenditure on pharmaceuticals in Zambia was estimated at 204 million USD in 2017 (BMI 2017), but the country has a serious problem with fake drug importation coming in from Asia despite the existing registration and regulation organ, Pharmaceutical Regulatory Authority of Zambia. These drugs are illegally imported by private drug sellers and distributed through the network of unlicensed private retail outlets, which further complicates establishment of reliable consumption estimates (AHO/WHO 2017).

WHOCC (2017) has reported the Defined Daily Doses (DDDs) for pharmaceutical compounds in order to collect data on the actual consumption of medicines. DDD is defined as “the assumed average maintenance dose per day for a drug used for its main indication in adults”. The global DDDs for the pharmaceuticals selected for this study are between 300 and 2000 mg, however the drug consumption volumes for Zambia alone are not available. The DDDs for the antiretroviral drugs are equivalent to the recommended doses: 300 mg for lamivudine, 400 mg for nevirapine and 600 mg for zidovudine. The DDDs for the antibiotics are 1000 mg for amoxicillin, ciprofloxacin and tetracycline, 100 mg for doxycycline, 800 mg for norfloxacin, 2000 mg for sulfamethoxazole and 400 mg for trimethoprim. All studied pharmaceuticals are also listed on the WHO’s Model List of Essential Medicines, which presents the most effective and safe medicines needed in a human health system (WHO 2017b). All studied drugs except norfloxacin are also listed on the Zambia Essential Medicine List (ZEML 2013).

Globally, nearly half a million people develop multi-drug resistant tuberculosis each year, and antimicrobial resistance has started to complicate the HIV treatment as well: an estimated 7% of people starting antiretroviral therapy in developing countries ended up having drug-resistant HIV in 2010. The increasing resistance might necessitate the use of more expensive second-line drugs in the future. This is problematic since majority of the people receiving ART live in the low- or middle-income countries, and cannot necessarily afford these new drugs (WHO 2017a). According to UNAIDS (2004), the Zambian AIDS-affected households have reported that their monthly income fell by 66-80% because of coping with HIV/AIDS-related illness. Most of these households were already poor, and the situation would likely worsen with the development of antimicrobial resistance in the HI-virus.

2.2 Agricultural use of urine

The agriculture and food system of Zambia is struggling with providing adequate nutrition and food security for all. Maize has become the staple food crop in Zambian diets after government subsidizing made farmers replace traditional food crops with it, and while part of the maize is produced through rain-fed traditional agriculture, inorganic fertilizers and other inputs are also used in maize production (Chizuni 1994; IIED & Hivos 2017). Soil fertility is in decline in southern Africa, which has also affected crop yields with growing food insecurity and environmental degradation in the region (Mafongoya et al. 2006).

With the increasing costs of inorganic fertilizers after the government subsidies and price control on maize and fertilizers were removed in order to compel farmers to switch back to crops that are better adapted to their region, many low-income smallholder farmers are not able to afford the fertilizers (Mafongoya et al. 2006; Chizuni et al. 1994). Especially women farmers are affected by the high cost and limited access to fertilizers all over sub-Saharan Africa (Andersson 2015), even with Zambia consuming more than double the amount of fertilizers than the average in the region (The World Bank 2017). Agricultural technologies that maintain the sustainability of resources and increase income generation and food security should be developed and supported (Chizuni 1994).

Ecological sanitation practices have the potential of using human excreta as a resource for nutrient recycling instead of seeing it as a hazardous waste for disposal and a source of pollution. Conventional approaches to sanitation, especially waterborne flush-and-discharge systems have, according to Windblad & Simpson-Hébert (2004), disrupted the naturally occurring cycles of life-building substances. In China, Japan and Europe, source-separating and using human excreta as a fertilizer has historically been a common practice, either separately or together with animal manure (ibid; Höglund 2001). Agricultural use of urine as a fertilizer has re-entered into the sanitation field during the last two decades in order to utilize the high macronutrient contents of human urine to meet the global sustainability and resource efficiency targets, especially in case of recovery of the finite raw material phosphorus (Kirchmann & Pettersson 1994; Jaatinen et al. 2016; Pronk & Koné 2009). Urine is often promoted as a cost-effective fertilizer that is seen as an environmentally sound and low-risk alternative, with social norms and cultural perceptions as the main constraints for the wider adoption of the practice (Andersson 2015). However, it is becoming more evident that in areas with high disease prevalence and high usage of medication, pharmaceutical traces can affect the safety of the measure (Jaatinen et al. 2016).

Urine is indeed a source of fertilizing nutrients as it contains macronutrients in liquid form, especially nitrogen N, phosphorus P and potassium K. Even though the nutrient contents of untreated urine are relatively low from the point of view of commercial fertilizers (0.9% N, 0.06% P and 0.3% K), especially phosphorus recovery is considered an advantage of urine fertilization, as phosphate is a finite resource with quality compromises and price increases in sight (Maurer et al. 2006). Up to 60-90% of the ingested plant nutrients can be recovered from urine, and urine is also the main source of nutrients in wastewater (Kirchmann & Pettersson 1994; Pronk & Koné 2009).

The human excreta produced annually by one person includes approximately 5.7 kg N, 0.6 kg P and 1.2 kg K, of which urine contains some 90% of the N, 65% of the P and 80% of the K (Wolgast 1993). Jönsson et al. (2004) analyzed annual excretion of nutrients in five countries, of which South African urine contained 3.0 kg of N, 0.3 kg of P and 1.2 kg of K and Ugandan urine 2.2 kg of N, 0.3 kg of P and 1.0 kg of K, respectively – Zambian values could perhaps be somewhere in between these two. In a study conducted by Kirchmann & Pettersson (1994), phosphorus contained by urine was at least as available to crops as soluble mineral P fertilizers. Heavy metal concentrations measured in the urine were low compared with other organic fertilizers. Nevertheless, Bischel et al. (2015) have measured high pharmaceutical concentrations in source-separated urine in eThekwini, South Africa. They found maximum concentrations of 6800 µg/L for the antibiotic sulfamethoxazole and 1300 µg/L for trimethoprim.

Urine can be applied to the crops either neat or diluted with water and either directly onto the soil or incorporated into it. Dilution levels vary from 1:1 to 1:10 and dilution has the perk of reducing the risk of over-application (Jönsson et al. 2004). Sene et al. (2012) recommend applying urine in frequent intervals instead of once before planting as well as using ash and compost together with urine to ensure good results. Windblad & Simpson-Hébert (2004) claim that the highest fertilizing effect is achieved when using urine in soils with high organic matter contents, which can be enhanced by adding garden compost or fecal matter from the UDDTs. Also the toilet wastes could be composted together, but the separating toilet can provide a better nitrogen economy as composting lowers the N content up to 70% (Heinonen-Tanski & Wijk-Sijbesma 2005).

Urine can be used for fertilizing a plethora of plants, including food plants. Fertilizing tests have been conducted with many vegetables such as tomatoes, beans, cabbage, beetroots, pumpkin, cucumber, okra and spinach (Kawanga et al. 2015; Ranasinghe et al. 2016; Pradhan et al. 2007, 2010a, 2010b; Heinonen-Tanski et al. 2007; Akpan-Idiok et al. 2012; Sene et al. 2012). In many cases the urine-fertilized plants yielded as good or better harvest and increase of biomass than similar plants fertilized with industrial fertilizers. In the peri-urban settlement of Madimba in Lusaka, urine was used for a test cultivation of tomatoes using drip fertigation in a greenhouse coupled with lime application. The urine was collected from 100 UDDTs built in the community (Picture 4.) and applied to the root zone of the tomato plants after a retention period of 3 months in a temperature of 20-30 °C. There was no leave contact, which might have damaged the plants. After a period of 24 weeks, the 160 m^2 greenhouse with 325 tomato plants yielded over 10 000 tomato fruits and therefore urine was seen as a good alternative to commercial fertilizers (Kawanga et al. 2015).

Picture 4. Urine is often collected through a pipe into a plastic container stored behind the separating toilet, and stored in similar containers for at least 6 months before application.

The fertilization potential of human urine has been studied elsewhere in sub-Saharan Africa as well. Akpan-Idiok et al. (2012) demonstrated that application levels of 20 000 liters of urine/ha in Nigeria increased the growth and yield of okra plants (*Abelmoschus esculentus*) compared to mineral fertilizer, while 15 000 liters of urine/ha produced similar yield of okra as the inorganic fertilizer. The urine was strongly alkaline and contained moderate amount of nutrients N, P, K, Mg, Ca and Na. A study conducted in Uganda by Andersson (2015) concluded that yields were improved and farmers elevated their skills after introducing the urine fertilization. The author states that using urine as a fertilizer is “valued as a low-cost and low-risk practice contributing to significant yield increases, suggesting important contributions to food security and income, especially for those who have few options in soil nutrient management.” This “low risk” refers to salt accumulation in soils, which Andersson (2015) considers not being a risk among African smallholders, but also to pathogens. Pharmaceutical residues were not taken into account, even though source-separated urine can contain significant amounts of such contaminants.

Winker (2009) states that non-degradable pharmaceuticals, such as many of the pharmaceuticals used for antiretroviral treatment (Jaatinen et al. 2016), can be incorporated by plants. Uptake of pharmaceuticals and other PPCPs can lead to these contaminants ending up in different parts of plants, also fruits of food crops, when irrigating or fertilizing the plants with materials containing such residues. Wastewater, sewage sludge, manure and urine are all potential sources for pharmaceuticals and other bioactive compounds when added to cultivated plants as soil conditioners (Eggen et al. 2011). Azanu et al. (2016) have found out that their potted test plants carrot (*Daucus corota L.)* and lettuce (*Lactuca sativa L.)* absorbed the antibiotics amoxicillin and tetracycline from water in all concentrations, most likely passively. Tetracycline was detected at concentrations up to 28.3 ng/g in lettuce and up to 36.8 ng/g in carrots, while amoxicillin concentrations were ranging up to 45.2 ng/g. Eggen et al. (2011) have also noticed uptake of ciprofloxacin in carrot (*Daucus carota* ssp. *sativus* cvs. Napoli) and barley (*Hordeum vulgare*). Pharmaceuticals were more concentrated in the roots than in the leaves of the studied plants, and they were negatively affecting the growth and development of the crops. Consuming low but constant antibiotic concentrations in food crops could lead to increased antimicrobial resistance (Azanu et al. 2016).

The direct fertilizer use of source-separated urine containing high amounts of non-degradable pharmaceuticals could also have effects on aquatic life and the resident microbial communities of soils and sediments (Kümmerer 2003). Richert et al. (2010) suggest that urine is better applied into the soil rather than to the conventional sewage system from where the pharmaceutical micropollutants would end up in the water bodies, because “the micro-pollutants can be degraded better in the aerobic, biologically active soil layers -- than in water bodies whose ecosystems are much more sensitive.” Jönsson et al. (2004) support this view, stating that the possible risk from pharmaceutical substances in the agricultural system is small compared to the waterborne system, and the human use of pharmaceutical substances is small compared to the amount of pharmaceuticals in animal manure and pesticides used in agriculture.

This may be relevant in the context of industrialized countries, but for example Bischel et al. (2015) have measured high concentrations of pharmaceuticals in source-separated urine in South Africa while Segura et al. (2015), K'oreje et al. (2016) and Ngumba et al. (2016) have found high concentrations of pharmaceuticals in groundwater, surface water and wastewater in Kenya and other countries in sub-Saharan Africa. In both last mentioned studies the highest concentrations were measured in close proximity to densely populated informal settlements where pharmaceutical residues enter the groundwater via on-site sanitation systems through soils and the surface waters often via direct discharges of wastewater. These findings suggest that the safety of urine fertilization would be best examined in relation to the local context including local medicine consumption patterns as well as local water and sanitation systems.

Because of the concerns on the uptake of pharmaceuticals and other possible risks related to using urine containing pharmaceutical residues as a fertilizer, urine from people under medication is not recommended for agricultural use (Winker 2009). According to Lienert et al. (2007), human metabolism reduces the toxicity of some pharmaceuticals, but unevenly: they studied some of the same antibiotics as in this study and found out that toxic potential of the parent drugs after metabolism is higher in the urine fraction than in the feces fraction in all except one studied antibiotic (norfloxacin). Therefore, urine source-separation could successfully decrease the ecotoxicological risks of certain pharmaceuticals. For instance Pronk & Koné (2009) suggest that pharmaceutical residues should be removed from the urine intended for fertilizer use in order to prevent long-term hazards.

The experience in the peri-urban settlement of Madimba in Lusaka is that the acceptance of the agricultural use of urine is depending on the local conditions and socio-cultural beliefs as well as on the status of legal recognition of this type of fertilizers in the local policy making. Kawanga et al. (2015) state that the people are facing "challenges in changing their mind set to implementing the full utilization of human urine", and this is partly because using human excreta as a fertilizer is not legally recognized in Zambia. However, if high levels of pharmaceuticals were found in the source-separated urine currently promoted for fertilizer use, its utilization could pose certain risks to human health and the environment (Pynnönen & Tuhkanen 2014). Preventive measures and careful management of the situation would in this case be required.

2.3 Pharmaceutical residues and the environment

Despite the beneficial fertilizing properties of urine discussed in Chapter 2.2, source-separated urine may contain pharmaceutical residues originating from human consumption, which, after ending up in the environment become environmental pollutants that are referred to as pharmaceuticals and personal care products (PPCPs) (Ngumba et al. 2016). These are classified in the wider group of Emerging Organic Contaminants (EOCs), and their adverse effects on aquatic life, animals and even human health are a growing concern despite their trace-level presence in the environment (Pal et al. 2010). The largest part of pharmaceutical substances ingested by people is finally excreted with urine, while a smaller portion is excreted with feces: the amounts vary between substances, but on average, approximately two thirds of consumed pharmaceuticals are excreted with urine and one third with feces (Richert et al. 2010). Also Lienert et al. (2007) found out that 64% (±27%) of the active ingredients in the studied pharmaceuticals were excreted via urine, and 35% (±26%) via feces. These pharmaceuticals can pose ecotoxicological and antimicrobial resistance risks when ending up in the environment (Jaatinen et al. 2016). In the peri-urban settlement of Madimba, this can happen either through the unsafe sanitation systems discussed earlier or through the agricultural use of urine containing pharmaceuticals.

Since the mid-1990’s, there has been a growing interest in pharmaceutical residues in the water systems among environmental science: a lot of knowledge has been produced concerning the situation in industrialized countries (Ngumba et al. 2016), but the matter is virtually non-studied in the urban water cycles of Zambia and many other developing countries. The occurrence and concentrations of pharmaceuticals used in antiretroviral treatment ART and their relationship with the environment are not extensively studied – antiretroviral drugs even less than antibiotics (Jaatinen et al. 2016). PPCPs in general are more investigated in the aquatic than in terrestrial ecosystems (Jjemba 2006). The occurrence of pharmaceutical residues in conventional wastewater treatment plants and their receiving water bodies in industrial countries has been studied, but these studies often take into account pharmaceutical compounds associated with high living standards such as painkillers, contraceptives and blood lipid regulators, but also antibiotics (Heberer 2002; Kümmerer 2009).

In industrialized countries the excreta is mainly flushed through the sewer network and thus the primary point sources that release pharmaceuticals into the aquatic environment have been identified as the wastewater treatment plants treating domestic, hospital and industrial wastewaters (Al Aukidy et al. 2012; Gulgowska et al. 2008; Pal et al. 2010). Other sources sources of EOCs are wastewaters from livestock and agriculture, while rivers can disperse EOCs to other water bodies, including aquifers, estuaries and marine systems. Direct discharge of wastewater to aquifers can occur through on-site (septic) wastewater treatment systems, which can pose a threat to groundwater supplies (Pal et al. 2010). Centralized water treatment and wastewater treatment plants serve only a fraction of the population in the developing world, where urban hydrological cycles are often based on on-site systems (Ngumba et al. 2016). This is also the case in the peri-urban areas of Lusaka, Zambia. In such areas without safely managed sanitation systems, a significant part of the consumed pharmaceuticals end up directly into the environment, either as unchanged parent compounds or as metabolic compounds (Jaatinen et al. 2016; Jjemba 2006).

The concentrations of PPCPs in the water bodies receiving wastewater effluents are typically low, in nanogram and microgram ranges in the industrial countries of the global North. Therefore the concentrations are not often acutely toxic to aquatic organisms, but the effects of long term exposure and chronic toxicity are still largely unknown (Jjemba 2006). The PPCPs are pseudo-persistent in aquatic environments due to their continual release from discharges of wastewater, either treated or untreated. These contaminants enter the wastewater mainly via urine and have potential of causing antimicrobial resistance and toxicity to aquatic organisms (Ngumba et al. 2016). The antibiotics used together with antiretroviral drugs for ART (such as CIP, TMP, TET, DOX and SMX) and their transformation products have been detected in conventional WWTP influent and effluents all over the world (Haddad et al. 2015; Gao et al. 2012). Also antiretroviral drugs are highly bioactive and they have been reported to be potential environmental pollutants (Jain et al. 2013). Moreover, the mixture or ‘cocktail’ effects of these EOCs can rarely be taken into account (Lienert et al. 2007).

One of the most important risks related to the pharmaceutical contamination of the environment is the potential antimicrobial resistance in pathogens, which can lead to development of resistant microbial populations and cause both health hazards and socio-economical problems. WHO (2017a) defines antimicrobial resistance as the “resistance of a microorganism to an antimicrobial drug that was originally effective for treatment of infections caused by it”, meaning that the medicine in question has lost its ability to destroy the microorganisms and thus heal the symptoms. Hence, antimicrobial resistance poses “an increasingly serious threat to global public health that requires action across all government sectors and society” (ibid). Antibiotics and antiretroviral drugs have the potential of spreading antimicrobial resistance in microorganisms through antibiotic resistance genes (ARGs) that are disseminated in the urban wastewaters and especially in the wastewater treatment facilities (Segura et al. 2015; Li et al. 2010). Also in natural environments, the continuing exposure to low doses of antibiotic substances can lead to the natural selection of resistant bacteria, and some concerns have been expressed that soil bacteria could develop antimicrobial resistance through such doses in groundwater (Archundia et al. 2017).

For instance, in a study conducted in Bolivia, Archundia et al. (2017) found that the studied antibiotic resistance genes were widely detected in the hydrological cycle and even in areas where antibiotic pollution was not detected. Bischel et al. (2015) also detected a sulfonamide antibiotic resistance gene in all studied source-separated urine samples in South Africa. According to WHO (2017a), resistance to pharmaceuticals used in HIV treatment has been increasing, and the matter requires urgent attention. Especially in areas with high disease prevalence and high environmental concentrations of pharmaceuticals, the risk of antimicrobial resistance development is acute and serious.

2.4 Treatment options for source-separated urine

In an on-site sanitation system, urine collection and treatment aims at preventing environmental pollution, hygienic risks and unpleasant smells, but also recovering nutrients (Udert et al. 2015). Source-separation of urine would be an effective measure for decreasing the ecotoxicological and other risks related to micropollutants such as pharmaceutical residues, as it would collect these substances into manageable volumes for further treatment or disposal (Lienert et al. 2007). If the source-separated urine containing pollutants should be designated for fertilizer use, it should be treated for sufficient hygienization and reduction of micropollutants (Maurer et al. 2006). Therefore, it would be essential to incorporate a safe treatment or disposal method in the sanitation plan utilizing urine-diverting toilets.

Larsen et al. (2004) present source control and source-separation as approaches to preventing the micropollutants such as pharmaceutical residues from entering wastewater. Source control is not a viable option in the study area, as despite the over-the-counter sales of pharmaceuticals that could be controlled by descriptions, the aim is rather increasing the access to medication than limiting it, especially among the populations living with HIV. As the population of the study area is not connected to sewer network or serviced by a wastewater treatment plant, source separation is the only viable approach for removal of pharmaceutical pollution from entering the environment directly or via wastewater. Urine source-separation has the advantage of protecting the environment from pharmaceutical contamination, as the majority of pharmaceuticals enter the environment through urine. Source-separation is also overall a more suitable measure for the complex task of dealing with micropollutants than end-of-pipe technologies – especially in settings with decentralized sanitation systems such as Zambia.

Currently WHO (2006) is recommending a storage period of ≥6 months in 20 °C for effective hygienization of source-separated urine before applying to the crops as a fertilizer. The WHO guidelines for the safe use of wastewater, excreta and greywater demonstrate that pathogens in source-separated urine are extensively destroyed after the suggested storage period of ≥6 months before applying to the crops, but this also depends on the storage conditions (ibid). This is supported by the results of Höglund et al. (2002), who found out that storage time, pH and especially temperature affect the pathogen inactivation rates, inactivation being more effective in 20 °C than in 4 °C. Maurer et al. (2006) have pointed out that there are many solutions for sterilization of any solution including heat, ultraviolet (UV) light and pressure, but as these have not been tested for urine, the storage may be the most appropriate and energy efficient.

Urine from unhealthy people may contain pathogenic microorganisms as well as prions, and urine can also get contaminated by feces during source-separation, which is why hygienization is a minimum prerequisite for its usage in agriculture (Maurer et al. 2006). For instance Schönning et al. (2002) have found a mean fecal contamination of 9.1±5.6 mg/L in the studied source-separated urine. According to Heinonen-Tanski & Wijk-Sijbesma (2005), urine should be diverted in the toilet structure to ensure that it is free from feces and related pathogens. Urine can be effectively hygienized by different treatment options, but even while fresh urine contains few enteric microorganisms, certain human parasites such as helminth eggs or *Schistosoma* can withstand the storage treatment and pose risks when applying urine of an infected person in agriculture. As these infections are common in the global South including Africa, this risk should be addressed. However, when comparing the risk with water contamination through pit latrines and open excretion, it has been estimated that the agricultural use of urine hardly constitutes an increased risk of disease transmission through the mentioned parasites (ibid).

Pharmaceutical compounds and other micropollutants do not necessarily degrade during a storage period. A study conducted by Jaatinen et al. (2016) shows that the residues of pharmaceuticals used in ART in the source-separated urine did not break down completely in the six month timeframe recommended by WHO (2006): the pharmaceutical concentration reductions were for antituberculotics 42-99%; for antivirals <52% (except for 3TC 76%) and for antibiotic compounds <50%. In assays performed with amendments such as feces mixed in the urine, the reductions were all less than 50%, despite the expected higher microbial activity. This means that after six months, the concentrations of pharmaceutical residues in urine had degraded only partially, on average to half of what the concentrations were in the beginning of the test. Also some pharmaceuticals were transformed into transformation products during the storage period, which can also have adverse effects. Schürmann et al. (2012) have had corresponding results, stating that pharmaceuticals in source-separated urine were eliminated only partially during storage, and such treatment method is insufficient for their removal. Also Bischel et al. (2015) found that 11 out of the studied 12 pharmaceutical compounds did not degrade to a significant extent during the storage period. These reduction rates may not be sufficient for the safe agricultural use of urine containing high amounts of pharmaceutical residues.

Besides storage, several more elaborate options for removal or degradation of the pharmaceutical residues are available, however they all come with their strengths and weaknesses (Maurer et al. 2006). The technological solutions that could be appropriate for the Zambian context are presented in the next paragraphs, with struvite precipitation, electrodialysis and evaporation as some of the potential technologies (Figure 4.) after Pronk & Koné (2009). Additionally, electrolysis and a combination of nitrification and distillation could be viable options in Zambia, as they have already been piloted in South Africa (Udert et al. 2015). By employing such techniques, source-separated urine could be processed into both solid and liquid fertilizer products that could enable safe usage of source-separated urine and perhaps provide local business opportunities. The aim would be to recover nutrients and increase their concentration, remove micropollutants and hygienize the end product (Pronk

& Koné 2009). Micropollutant removal processes are generally based on oxidation or adsorption whereas separation processes are mainly based on membranes or precipitation (Maurer et al. 2006). These technologies could often involve high costs, which is why the appropriateness of the applied technologies should be evaluated not only by effectiveness but also by the treatment costs. The concentration of nutrients and volumes is important for choosing the treatment process as transport and storage may also induce continuous costs. According to Pronk & Koné (2009), it is important to ensure that the urine collection and treatment approach is market-driven and takes the added value into account. Udert et al. (2015) have estimated that any process of urine treatment will be implemented on a large scale only “if the investment and operational costs are low and if the treatment technologies are reliable and easy to operate.”

Perhaps the most extensively tested option for treatment of source-separated urine is the production of magnesium ammonium phosphate ($MgNH_4PO_4 \cdot 6H_2O$), also called struvite. It is a solid fertilizer that is formed by the addition of magnesium to urine, usually in the form of MgO, $Mg(OH)_2$, $MgCl_2$ or bittern, which is the Mg-rich brine from table salt production (Pronk & Koné 2009; Maurer et al. 2006). Also wood ash could be used, but it could induce challenges with heavy metals and precipitation of unwanted minerals (Udert et al. 2015). Struvite is a white powder that is rich in N, Mg and P and can be used as a slow-release fertilizer. In the relatively simple and fast process, source-separated urine is first precipitated with MgO and washed with saturated struvite solution. Then it is dried at 30 °C, after which urine has been processed into a powder that is free of pharmaceuticals and pathogens (Schürmann et al. 2012). No pH adjustment is needed as the pH of hydrolyzed urine is suitable for struvite precipitation. Other nutrients, such as 97% of the nitrogen and practically all of potassium and sulphur remain in the effluent, but struvite precipitation can be combined with ion exchange for better nitrogen recovery by adding zeolite to the urine solution (Udert et al. 2015; Maurer et al. 2006). As a solid fertilizer, struvite allows for easy transportation and the technology has been tested in two pilot projects in Asia (Antonini et al. 2011; Etter et al. 2011) and in South Africa, also with manually operated reactors (Udert et al. 2015). However, all the micropollutants and some pathogens remain in the liquid phase that is collected as a supernatant after the precipitation, and therefore struvite precipitation needs to be combined with wastewater treatment processes for the effluent in order to prevent environmental pollution and hygiene risks (Pronk &

Koné 2009; Udert et al. 2015). In addition, a magnesium source is needed for the precipitation process, which also adds to the costs (Pronk & Koné 2009).

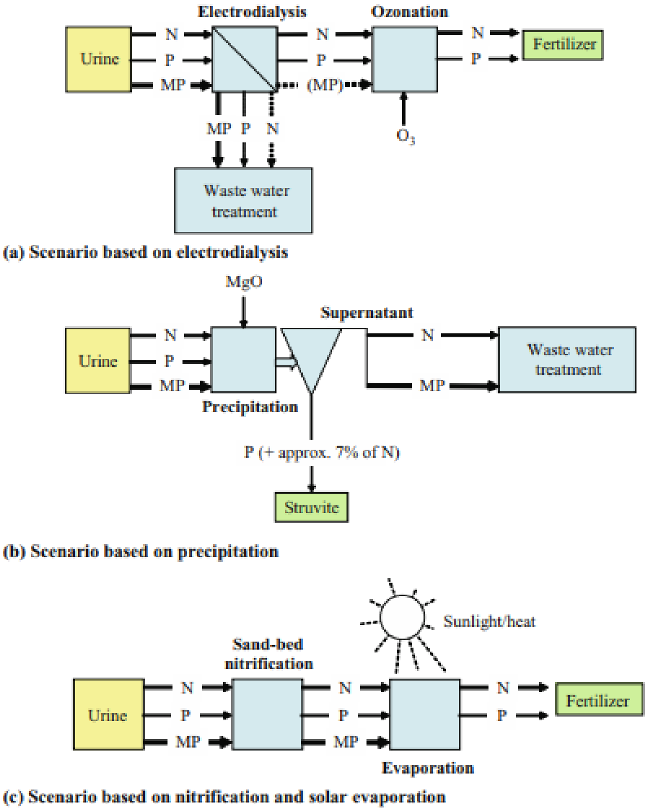


Figure 4. Electrodialysis, struvite precipitation and nitrification and solar evaporation are scenarios for production of fertilizers from source-separated urine in a developing country context (Pronk & Koné 2009).

Application of electrodialysis, where ions permeate through positively and negatively charged membranes and concentrate in a separate compartment, has been investigated on laboratory and pilot scale for the treatment of urine (Pronk & Koné 2009). Bacterial hygienization is based on the electrodialysis membrane, which is an ion-exchange membrane with a dense structure and pore size of <1 nm, and the removal of micropollutants is jointly based on adsorption to the membrane and on the sieving effect (Pronk & Koné 2009; Maurer et al. 2006). Pronk et al. (2006) have investigated electrodialysis for the selective separation of nutrients from source-separated urine and found out that all the nutrients present in urine can be concentrated by a factor of 1.6 to 4.1. The studied spiked pharmaceuticals, none of which the same ones as in this work, were effectively removed initially, but breakthrough of some compounds occurred after extended operation times (approximately 400 days). Ozonation would be needed to remove these remaining micropollutants. Electrodialysis has several other limitations as well. The wastewater from the process still contains considerable amounts of nutrients and micropollutants, and would need treatment. Also the investment and energy costs generated from the membrane and other equipment as well as from the energy consumption of approximately 97 kWh/m^3 pose a limit to application of electrodialysis in developing countries (Pronk & Koné 2009).

In addition, Udert et al. (2015) suggest that electrolysis, a process based on indirect oxidation, could be utilized in small on-site reactors which could be integrated into toilets, cutting on the transportation costs. High degradation rates per surface area could be achieved with electrolysis, and the process is simple to operate. It would possibly produce only small amounts of by-products such as struvite, and the remaining water could be infiltrated. The drawbacks of electrolysis are high usage of electricity and high prices for the most efficient electrodes. Also nitrogen is not recovered in the process and chlorinated by-products are formed, which can be hazardous for human health. Electrolysis could be suitable if environmental protection is a more important goal than nutrient recovery.

Evaporation is a technology for urine volume reduction by removal of water. Pilot tests have been carried out on a laboratory scale so far, when fresh urine was evaporated in a pressure of 200 mbar and a temperature of 78 °C, resulting in a tenfold volume reduction and an end product of a viscous liquid that contained 9.7% nitrogen (Maurer et al. 2006). Sand-bed nitrification and solar evaporation could be an option for Zambian context as sunlight is abundant in the country, but this technology presented by Pronk & Koné (2009) is yet to be fully developed. The ammonia contained by urine should be first stabilized with acidification or nitrification, the latter being a more viable option. An intermittent sand filter or vertical flow constructed wetlands have achieved a nitrification efficiency of up to 90%, which could be suitable for low-cost applications. In this system, the nitrate could be collected with the effluent and concentrated for further usage by solar evaporation. The UV radiation degrades micropollutants and pathogens effectively (Pronk & Koné 2009).

Nitrification in sand filters and solar evaporation is a potentially viable option for developing countries, as it is a low-cost solution that would also enhance the development of local business. The main limitation is the potential of denitrification of the end product, but the process needs further development and testing specified for urine applications (Pronk & Koné 2009). Also the nitrate production is limited, since the process stops due to the low pH when half of the ammonia is oxidised to nitrate, and alkalinity should be added to achieve a better conversion rate (Udert et al. 2015). A combination of nitrification and distillation could also be an option that would allow for recovery of all nutrients without addition of chemicals. This process combination recovers nutrients into one concentrated and hygienized solution through nitrification, and concentrates the nutrients in low volumes by distillation, producing only distilled water and a small amount of sludge as by-products. Udert et al. (2015) have piloted the process in a research project in eThekwini, South Africa.

Ozonation can be used for elimination of a wide range of micropollutants from urine. It is based on the addition of chlorine, chlorine dioxide, ozone or OH radicals that are reactive with most organic micropollutants. Laboratory tests have demonstrated complete oxidation of certain micropollutants, including pharmaceuticals. Adsorption to active carbon or other adsorbents could be an additional option for micropollutant removal as well, but the high amounts of pollutants causing a high COD in urine would most probably interfere with the process. In addition, micropollutant separation rates of over 92% could be achieved with nanofiltration in optimized conditions, but the efficiency of the process depends strongly on the pH. Also ammonia stripping has proved effective for the separation of nutrients and micropollutants (Maurer et al. 2006). Nevertheless, the implementation costs of these four technologies are probably a limitation for applications in developing country contexts.

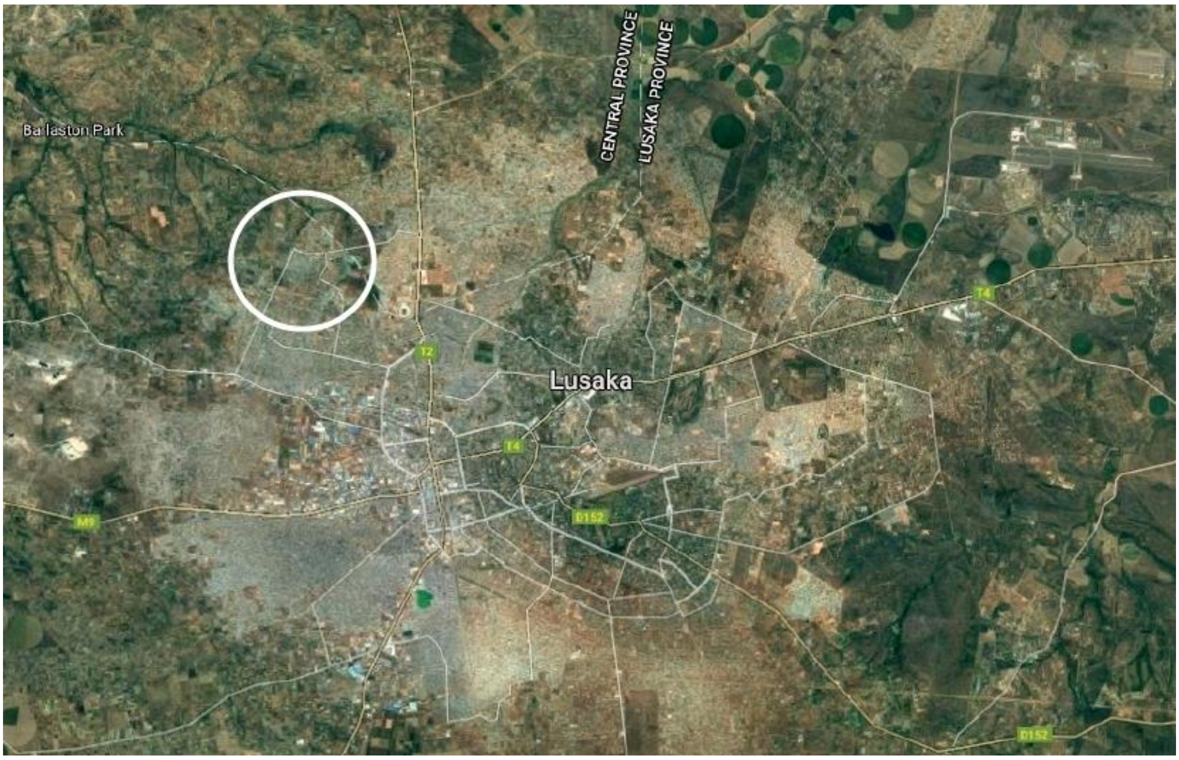
3 MATERIALS AND METHODS

In order to study the occurrence of antibiotics and antiviral drugs in the peri-urban area of Madimba in Lusaka, empirical quantitative data needed to be obtained. Water and urine samples were collected and prepared with solid phase extraction (SPE) during the data collection period in Lusaka in May-July 2016. The extracted samples were shipped to the University of Jyväskylä for analysis with liquid chromatography-tandem mass spectrometry (LC-MS/MS). Also observation and interviews were used for collection of qualitative data. This chapter describes the study area, the used sampling method, sample preparation and analysis methods as well as the chemicals and standards used. Also the complementary qualitative data collection methods will be explored.

3.1 Description of the study area

The peri-urban community of Madimba is situated some 10 kilometers northwest from the Lusaka city center as presented in Picture 3, between the large low-income housing area of Chunga and the peri-urban farming area Barlaston Park. Madimba is an informal, unplanned settlement with an approximate land area of 1.2 km². The settlement has been inhabited from the year 1960, when it was first occupied by four households and a group of American missionaries (Kawanga 2005). According to the Global Dry Toilet Association of Finland GDTF (2017a), the population of Madimba has doubled since 2005 and today the community is home to some 6000 people, of which 47% are male and 53% female (Kawanga 2005). Most of the adult population is monogamously married and the majority is renting their accommodation (ibid).

Approximately 20% of the inhabitants have small gardens for subsistence farming and income (GDTF 2017a). In fact, Madimba is a local name that means gardens or farms (Kawanga 2005). Kawanga et al. (2015) estimate that there are more than 200 women practicing urban agriculture in Madimba with rented farm land ranging from 600 to 1200 square meters in size. The crop yields are below sustenance levels and inadequate to achieve food security. Based on the field observation notes, some of the residents also sell their produce in the local stores or by the road as a livelihood.



Picture 5. The peri-urban community of Madimba is located 10 km northwest from the center of Lusaka as indicated by the white circle (Google 2017).

The area does not have a sewage network or a widespread water supply network (GDTF 2017a; Kawanga 2005). The groundwater quality and levels fluctuate seasonally, and especially during the rainy season, latrine wastes are spread in the environment with the flooding water that soaks the yards and roads because of the inadequate ditch and drainage systems (GDTF 2017a). According to Kawanga (2005), the shallow wells become more contaminated with fecal coliforms during the rainy season. As Madimba was long lacking the service of the licensed water supply provider, many residents have dug shallow groundwater wells in their yards and use this well water for their daily chores. These wells are the main source of water in Madimba, accounting for 68% of the household water usage according to Kawanga (2005). Furthermore, public communal taps with monthly cost-sharing fees accounted for 20% of water usage and 12% of the water originated from other sources. The residents relying solely on communal taps often have to walk up to a kilometer to reach the water point (ibid).

Many of the shallow wells are unprotected, but some protected wells also exist. These, after Grönwall et al. (2010), are covered with a lid and “protected from runoff water by a well lining or casing that is raised above ground level and a platform that diverts spilled water away from the well.” As described also by Kawanga (2005), the openings of some wells are protected against flooding, surface water, animals and solid waste from entering the well, usually with concrete structures of 10-12 cm of height, while wells with openings on ground level are more exposed to such sources of pollution. Nevertheless, all shallow wells are vulnerable to contamination occurring in the groundwater aquifer.

The depth of the shallow wells varies from less than two meters to more than five meters, depending on the height of the groundwater table that varies in different parts of Madimba. Despite the small size of the community, the area consists of three different hydrogeological zones, the groundwater table being particularly high in the southern and eastern parts of Madimba (Kawanga 2016). The waterlogged terrain covers approximately 40% of the land area in the community. The benefits of a plentiful water source are compromised due to contamination especially in these parts (Kawanga 2005). However, based on the sample background data, it seems that many residents of Madimba use the unsafe shallow well water only for washing and cleaning purposes, but some also drink it. Grönwall et al. (2010) support this observation by stating that the water sources in the low-income settlements of Lusaka vary between settlements and between different parts of the settlements. The preferred water source also depends on the intended use of the water and on the adequateness and reliability of the water source. Drinking water is often taken from a narrower range of sources for quality reasons.

When the sanitation development projects started in the area, there was only one tapped water source (GDTF 2017a). Recently the water supply network of the Lusaka Water and Sewerage Company has been expanding in the area, as some residents have been joining the water supply network based on the sightings of water taps on the yards during the data collection for this research. In addition, a few houses possess private borehole wells with water tanks but these are rare. In sum, while tap water connections are becoming more common, the majority is continuing to use hand-drawn shallow well water for household purposes and some even for drinking.

The sanitation facilities in Madimba are mainly pit latrines as in other peri-urban settlements (Kawanga 2005; Yasini 2007), but currently there are also several urine-diverting dry toilets (UDDTs), which can be also referred to as Ecosan toilets or urine diversion dehydrating toilets. Also improved VIP latrines have been constructed and retrofitted during the dry sanitation projects taking place in the area since the year 2008 with the aim of improving the sanitation situation of the community and decreasing groundwater pollution by preventing the pathogens from leaking into the groundwater from the pit. Also some households have constructed private septic tanks with water closets. This is supported by the findings of Yasini (2007) describing the peri-urban areas of Lusaka in general. During the Baseline survey (Kawanga 2005), it was found out that safe water was the greatest concern in Madimba community, with one in four respondents mentioning it. Lack of drains accounted for 9% of the respondents and lack of toilets was a concern in only 2% of the responses.

Picture 6. In densely populated areas the shallow wells are often located in a close proximity to pit latrines, which can be seen on the background.

As in other informal peri-urban areas, the water and sanitation situation is problematic also in the community of Madimba. The use of pit latrines is common in the low-income settlements of Lusaka, but the frequent use of them forms a threat to groundwater quality through contamination not only in the immediate environment but also in a larger range of the city, since the low-income settlements of Lusaka are often situated over the aquifer recharge areas (Nkhuwa 2006). In Madimba, the insufficiently constructed, numerous pit latrines are contaminating the groundwater table that is particularly high and close to the surface in parts of the study area (GDTF 2017a). The pit latrines are also constructed in close proximity with the groundwater wells as shown in the Picture 6, which allows percolation of contaminants through the soil into the groundwater, this being the major environmental concern associated with pit latrines (Yasini 2007). According to Kawanga (2005) and GDTF (2017a), many wells have already been contaminated with fecal pathogens such as *E. Coli* and *Salmonella* through the fecal-oral transmission route, which has led to diarrheal cases and several cholera epidemics occurring in Madimba.

The study area hosts one of the seven conventional wastewater treatment plants of Lusaka, the trickling filter wastewater treatment plant of Chunga, and a non-convectional wastewater treatment pond system. The latter, Matero wastewater stabilization ponds, are currently operating with limited functionality due to ageing and lack of adequate maintenance. These wastewater stabilization ponds in Lusaka have limited functionality due to several other factors that are shortening the hydraulic retention time, including sludge accumulation in and erosion of the ponds (Brown et al. 2012).

3.3 Samples

A total of 31 water samples and 10 urine samples were collected in the study area in June-July 2016. The water samples included 26 groundwater samples from 21 shallow wells and 5 borehole wells, as well as 5 surface and wastewater samples.

Together with each sample, a background information questionnaire form was filled. There were separate forms for water samples and urine samples (Appendices 1 and 2). The form was designed in Finland in spring 2016, before doing the data collection trip to Lusaka, and modified while on the field, based on the local conditions and availability of information that were observed during the finalization of the sampling plan. Some of the information obtained for the background questionnaire were directly estimated or measured by the researcher, such as the proximity of a well to a pit latrine, depth of a well or the turbidity of the sampled water and urine. However, for obtaining other background information, the local residents had to be interviewed. They were often the owners of the UDDTs or the wells, or their family members. The questions included for instance the number of users of the well or the daily medication patterns in the household. Location of each sample was also measured with a GPS tracker and a map was constructed based on these (Picture 7). Photographs were also taken from the location and surroundings.

Picture 7. Map with GPS locations of all the taken samples in the peri-urban areas of Madimba and Chunga in northwest Lusaka, Zambia (Google 2017).

3.1.1 Sampling plan and method

The sampling process was initiated with establishing a sampling plan for collecting the water and urine samples. First a certain number of toilets and wells had to be chosen for sampling. A ready-made list of the constructed urine-diverting dry toilets (UDDTs) in the study area was provided by NECOS and it was used for verifying and mapping the toilets where sampling was possible to carry out. For the groundwater wells, a list had to be composed first by exploring the study area. All the suitable toilets and wells found in the study area were placed on a map by their GPS coordinates which were taken during the exploratory phase.

A total of 69 groundwater wells were mapped and visited by walking around and exploring the community of Madimba. Due to limited time and resources, the composed list was not exhaustive and therefore not covering all the wells in the area, but it gave a good selection of wells to be chosen for sampling. Also the geological location of wells was taken into account: the study area consists of three geologically differing zones, and it was ensured that wells were chosen comprehensively throughout the area from each zone. Therefore, the criteria for the selection of the wells were: 1) convenience; 2) geological location; 3) representativeness of protected and unprotected wells.

From the mapped groundwater wells, a sample was chosen for water sampling by using the convenience sampling method, which is a non-probability sampling technique based on convenience and accessibility. It is also called accidental, haphazard or opportunity sampling as the subjects may be selected in the sample because they are situated, by accident, where the researcher is collecting data. In convenience sampling, the researcher chooses a sample of target subjects or units from a population or universe. These are included for the purpose of the study and because they meet “certain practical criteria, such as easy accessibility, geographical proximity, availability at a given time, or the willingness to participate.” Convenience sampling is often used in quantitative research for pilot testing and collection of biological and ecological data in the field, such as along roads or trails or in areas where subject density is high (Etikan et al. 2015). The results drawn from groundwater wells chosen by convenience sampling probably do not differ from the scenario where the wells would have been chosen by random sampling, as the conditions are similar and the aquifer is in risk of pollution throughout the study area.

Urine samples were chosen to be collected from public UDDTs of Madimba for ethical reasons in order to guarantee the anonymity of the samples, as any kind of exposure to stigmatization was avoided. In this case, the definition of public was extended to include also shared private UDDT facilities used by more than one household and more than 10 people, because there were only a few actual public toilets. After visiting the toilet owners on the NECOS’s list, 10 suitable toilets were found in the study area, some being in need of minor repair works before sampling could be carried out. As the number did not exceed the planned number of samples, no further selection method was implemented. The criteria for the selection of toilets for urine sampling were: 1) used by more than one household; 2) used by more than 10 people; 3) urine separation and collection is in operation or can be enabled with minor repairs.

3.1.2 Groundwater samples

A total of 26 groundwater samples were collected from the study area in June 2016. 21 groundwater samples were collected from protected and unprotected shallow wells and 5 samples from borehole wells (Picture 8). All the collected samples were grab samples. USEPA (2004) defines a grab sample as a “sample collected at a particular time and place”, which is representative of “only the composition of the source at that time and place.”

Picture 8. The groundwater samples were collected from 21 shallow wells and 5 borehole wells in the study area. The looks and locations of the wells were varying.

Duplicate 600 mL grab water samples were collected from groundwater wells according to the sampling plan. The groundwater samples from shallow wells were taken from the buckets which the well water was normally drawn with, by pouring the water into the cleaned and dried PET bottles and closing with the cap. The bottle was first rinsed with the well water. Two shallow well water samples were taken directly from the well, from a depth of approximately 10 cm. The borehole samples were collected through the connected taps or hoses. After collection, the samples were transported on ice and stored in a refrigerator at +4°C and protected from light while awaiting extraction within one week. Appropriate protective clothing was used while sampling.

3.1.3 Surface and wastewater samples

A total of five surface and wastewater samples were collected in July 2016. The duplicate 600 mL surface and wastewater grab samples were taken from Chunga River and the wastewater treatment facilities discharging their waters into the same river (Picture 9). The two surface water samples were collected from the Chunga River, one of these sited being the Chunga wastewater treatment plant effluent discharge site, while the three wastewater samples were collected from Matero wastewater stabilization ponds.

Picture 9. Surface water samples were collected from Chunga River while wastewater samples were collected from wastewater stabilization ponds.

The Chunga WWTP is a conventional, trickling filter wastewater treatment plant that discharges its effluent into the Chunga River. The surface water sample B was taken from the river, some two meters from the WWTP effluent discharge point, while the sample A was taken from the same river under a road bridge, towards upper reaches from the WWTP. Both locations had large amounts of solid waste floating in the river. The Matero wastewater stabilization ponds are a non-conventional wastewater treatment system, which are currently not operating in their full capacity due to ageing, lack of adequate maintenance, sludge accumulation and erosion (Brown et al. 2012). One sample was taken from an overgrown stabilization pond, while two samples were taken from the influent and effluent sides of another pond that visually gave an impression of being in a better state.

A total of 5 surface and wastewater samples were collected directly from the water body from the depth of 1-2 cm. All the samples were collected into food-quality, cleaned and dried PET bottles with a volume of 600 mL except for one sample with a volume of 500 mL. After collection, the samples were transported on ice and stored in a refrigerator at +4°C and protected from light while awaiting extraction within one week. Appropriate protective clothing was used while sampling.

3.1.4 Urine samples

The duplicate 125 mL urine grab samples were collected from public urine-diverting dry toilets of the study area. Some of the samples had been in storage for several months, some of these being hygienized because of future application as a fertilizer and some because the urine separation of the toilet was not working properly. In addition one sample was provided in a separate container. A total of 10 urine samples were collected.

The samples were poured from the urine containers into cleaned and dried food-quality glass jars with a volume of 125 mL and closed with a metallic lid (Picture 10). One sample was fetched from a container that could not be removed from its place by using a plastic bag. After collection, the samples were transported on ice and extracted immediately afterwards. Appropriate protective clothing and equipment was used while sampling.

Picture 10. Samples of source-separated urine were collected from public urine-diverting dry toilets of the study area.

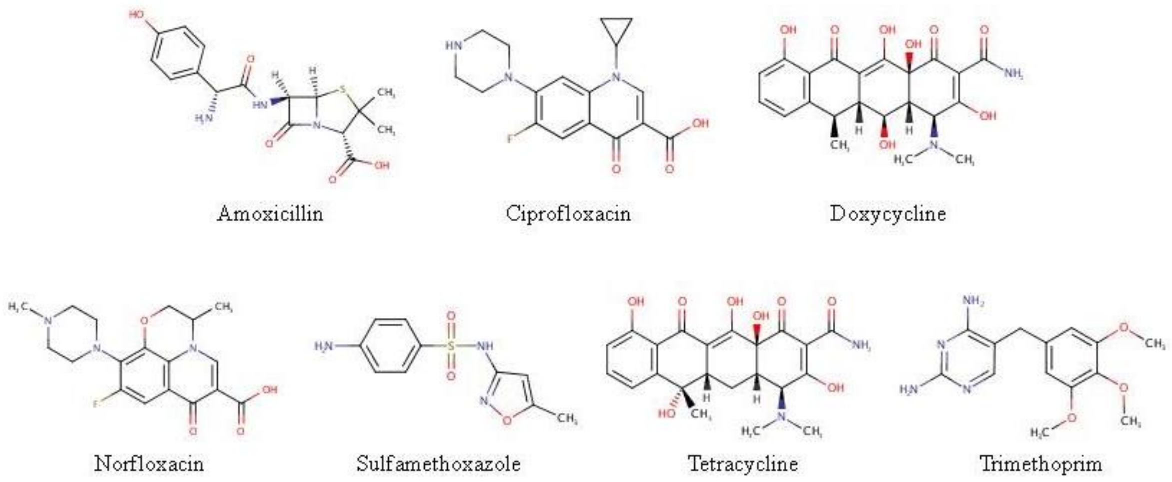
3.2 Chemicals and standards

In this chapter, the selected pharmaceuticals are presented together with their chemical and environmental properties. Also the internal standards and other chemicals that were used in the sample preparation are described.

3.2.1 Selected pharmaceuticals

The selection of the pharmaceutical compounds for this study is based on the criteria of plausible consumption of medications in the study area caused by the heavy disease burden, especially the high HIV prevalence and large amount of ART users as presented in Chapter 2.1.4. Also experience from the previous experiments of the supervisor and interest for environmental protection concerning the toxicity, pseudo-persistency and non-biodegradability of the compounds was of importance when selecting the pharmaceutical compounds (Jaatinen et al. 2016). Other affecting factors are impacts for human health concerning possible development of antimicrobial resistance (WHO 2016) and availability of detection techniques (Ngumba et al. 2016).

Amoxicillin (Figure 5.) is one of the most commonly prescribed antibiotics globally and is often detected in WWTPs (Toxnet 2017; Andreozzi et al. 2005). Less than 30% of amoxicillin is biotransformed (Drugbank 2017) and approximately 60% of the ingested amount of AMO is excreted in the urine, most of it unchanged (Rolinson 1973; Drugbank 2017). The main degradation product is amoxicilloic acid (AMA) (Toxnet 2017). AMO has a low distribution coefficient log K_{ow} (octanol-water partition coefficient) at 0.87 which indicates that the compound is highly mobile in soil (Kim et al. 2012).



Amoxicillin Ciprofloxacin Doxycycline

Norfloxacin Sulfamethoxazole Tetracycline Trimethoprim

Figure 5. Chemical structures of the selected antibiotics (Toxnet 2017).

Table 1. Properties of the selected antibiotic and antiretroviral pharmaceuticals (Drugbank 2017; [a]Le-Minh et al. 2010; [b]Kodešová et al. 2015; [c]WHOCC 2017; [d]Toxnet 2017; [e]Török et al. 2009).

	Properties of selected pharmaceuticals					
Compound	CAS reference	Molecular weight (g/mol)	Water solubility (mg/mL)	Log K_{ow}	DDD (mg)[c]	Excretion rate (%)
AMO	26787-78-0	365.4	0.96	0.87	1000	60
CIP	93107-08-5	331.3	1.35	1.313[a]	1000	50
DOX	564-25-0	444.4	0.63	−0.540[a]	100	20-26[d]
NOR	70458-96-7	319.3	1.01	1.478[a]	800	25-40
SMX	723-46-6	253.3	0.46	0.89[b]	2000	54[d]
TET	60-54-8	444.4	1.33	−1.470[a]	1000	40-55[d]
TMP	738-70-5	290.3	0.62	0.9[b]	400	50-60
3TC	134678-17-4	229.3	2.76	−1.4	300	86[d]
NVP	129618-40-2	266.3	0.11	2.5	400	80[e]
ZDV	30516-87-1	267.2	16.30	0.05	600	75

Ciprofloxacin (Figure 5.) is a broad-spectrum antimicrobial with an excretion rate of 40-70% after administration through the human body as unchanged compound (Drugbank 2017; Pal et al. 2010). Four metabolites of CIP (desethyleneciprofloxacin, sulfociprofloxacin, oxociprofloxacin, and N-formylciprofloxacin) have been identified in human urine, accounting for approximately 15% of an oral dose (Drugbank 2017). Ciprofloxacin has been detected in surface waters and wastewater effluents (Toxnet 2017), but it is expected to be photodegradable in sunlight and immobile in soil with a log K_{ow} of 1.313 (Le-Minh et al. 2010). Biodegradation is not an important environmental fate process in soil or water and the compound does not volatilize (Toxnet 2017).

Doxycycline (Figure 5.) is one of the synthetic antibiotic and antiparasitic drugs in the tetracycline derivative class (Drugbank 2017). DOX is very slightly soluble in water and sparingly soluble in alcohol. It is reasonably anticipated to be a human carcinogen (Toxnet 2017). 40% of DOX is excreted in the urine (Drugs.com 2017). The drug is not readily biodegradable (Drugbank 2017) and it has a log K_{ow} of −0.540 (Le-Minh et al. 2010).

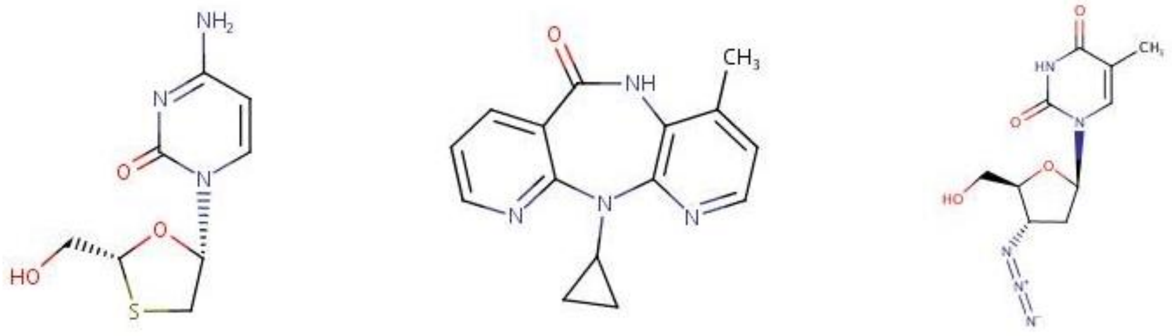
Norfloxacin (Figure 5.) is a synthetic antibacterial of the fluoroquinolone class (Drugbank 2017). Approximately 25-40% of the norfloxacin dose is excreted in urine as unchanged drug and 5-10% as metabolites. The drug has six metabolites, some of them being microbiologically active, but less active than the parent drug. The solubility of NOR depends on the pH and temperature. The drug is soluble in aqueous solutions with pH between 2 and 5 and over 9, and sparingly to slightly soluble in aqueous solutions with pH 7 (Toxnet 2017). Norfloxacin is immobile in soil with a log K_{ow} of 1.478.

Sulfamethoxazole (Figure 5.) is an antibiotic belonging to the class of sulfonamides (Drugbank 2017). Sulfamethoxazole is metabolized into several metabolites. The findings on the recovery vary, but approximately 10-20% of the ingested dose of SMX is excreted as unchanged parent compound, while 40-80% is excreted as the acetylated derivative, 15-20% as the glucuronide conjugate and 2-5% as hydroxylamine. 39-54% of the ingested dose is excreted into urine (Toxnet 2017; Pal et al. 2010). SMX is practically insoluble in water (Toxnet 2017), but in soil, SMX is expected to have high mobility based on its weak sorption properties with an estimated K_{oc} of 72 and a log K_{ow} of 0.89 (Deng et al. 2016; Kodešová et al. 2015). The compound is hydrolytically stable with pK_a values of 1.7 and 5.6, and not readily biodegradable, therefore persisting in soils (Kodešová et al. 2015). In the aquatic environment, sulfamethoxazole is not expected to adsorb to suspended solids or sediments and it is degraded by photodegradation (Toxnet 2017).

Tetracycline (Figure 5.) is a broad-spectrum polyketide antibiotic (Drugbank 2017). Approximately 40-55% of the ingested dose is excreted into urine (Toxnet 2017). Tetracycline is not metabolized but it is excreted in a biologically active form (Drugbank 2017).

Carlsson et al. (2006) have estimated that 100% of tetracycline enters the sewage in a waterborne sanitation system. Tetracycline has a log K_{ow} of −1.470 (Le-Minh et al. 2010).

Trimethoprim (Figure 5.) is an antibacterial agent related to pyrimethamine, of which 50% to 60% is excreted in the urine, mostly (80-90%) as unmetabolized parent compound (Drugbank 2017). Some 4% of TMP is excreted into feces and the rest is excreted as inactive metabolites (Toxnet 2017). Pal et al. (2010) have estimated a higher excretion rate of 70% for TMP. TMP has a pK_a of 7.12 and log K_{ow} of 0.9 (Kodešová et al. 2015).



Lamivudine

Nevirapine

Zidovudine

Figure 6. Chemical structures of the selected antiretroviral drugs (Toxnet 2017).

Lamivudine (Figure 6.) is a synthetic nucleoside analogue medicine with a short elimination half-life (Török et al. 2009). 70% of the ingested dose is excreted in urine as unchanged parent compound (Kumar et al. 2006). The only known metabolite of lamivudine is the trans-sulfoxide metabolite, which covers approximately 5% of the excreted compounds. The compound is immobile in soils with a log K_{ow} value of -1.4 (Drugbank 2017).

Nevirapine (Figure 6.) is a non-nucleoside reverse transcriptase inhibitor belonging to the class of alkyldiarylamines (Drugbank 2017). A small fraction of the dose (2.7%) is excreted in urine as the parent compound (Riska et al. 1999) and 80% of the compound and metabolites are excreted into urine and 10% are excreted into feces (Török et al. 2009). According to Cammett et al. (2009), the 2-, 3-, and 12-hydroxy metabolites of NVP were excreted in the urine at approximately 23%, 33%, and 30%, whereas the 8-hydroxy and 4-carboxy metabolites of NVP showed minor excretion rates, at 3.2% and 0.7%. NVP is also photostabile and relatively persistent in soils, but it can be biodegraded by aerobic microorganisms (ibid). It has a log K_{ow} value of 2.5 (Drugbank 2017).

Zidovudine (Figure 6.) is a potent nucleoside reverse transcriptase (Drugbank 2017). ZDV (or AZT) is a prodrug that is phosphorylated into its monophosphate, diphosphate, and triphosphate forms within the cell (Harlass 1996). The active metabolite is the zidovudine triphosphate form (Drugbank 2017). 45% of the zidovudine is excreted into urine as its major inactive metabolite, while approximately 15-29% of the dose is excreted unchanged (Drugbank 2017; Harlass 1996). ZDV is potentially carcinogenic to humans (Toxnet 2017) and it has a log K_{ow} value of 2.5 (Drugbank 2017).

3.2.1 Internal standards and other materials

Internal standard pharmaceuticals with a purity of ≥95% were added to the samples for identification and confirmation of the target analytes. The internal standards of Amoxicillin and [$^{2}H_{9}$]-Trimethoprim were purchased from Sigma-Aldrich (Steinheim, Germany), whereas the internal standards [$^{2}H_{8}$]-Ciprofloxacin, [$^{2}H_{4}$]-Sulfamethoxazole, [^{13}C, $^{2}H_{3}$]-Zidovudine, [$^{2}H_{4}$]-Nevirapine and [^{13}C $^{2}H_{2}$ $^{15}N_{2}$]-Lamivudine were purchased from Alsachim (Illkirch, France). All the remaining pharmaceutical standards were a kind donation from Universal Corporation Ltd, Kenya.

Individual standard solutions were prepared at a concentration of 1000 mg/L. Apart from ciprofloxacin which was dissolved in ultrapure water; all the other compounds were dissolved in methanol. The standards were subsequently diluted with 1:1 (v/v) methanol/ ultrapure water to a pooled mixed standard of 10 mg/L as stock solution and stored at +4°C in the dark.

Glass microfiber filters with a size of 47 mm and retentions GF/D (2.7 µm) and GF/F (0.7 µm) were purchased from Whatman (Maidstone, England). Oasis hydrophilic-lipophilic balanced (HLB) solid phase extraction (SPE) cartridges (6 mL, 200 mg and 3 mL, 60 mg) were obtained from Waters (Milford, USA). Analytical grade methanol was obtained from EMSURE® (Merck KGaA, Darmstadt, Germany). Unless otherwise indicated, all the chemicals used in the study were of analytical grade or above.

3.3 Sample preparation with SPE

Solid phase extraction (SPE) was used for extraction, concentration and clean-up of the water and urine samples which were afterwards transported to University of Jyväskylä for analysis.

The duplicate 20 mL urine samples were directly passed through Oasis HLB cartridges (3 cc/60 mg) by using a 20 mL plastic syringe (Picture 11). The urine was then disposed of into a septic tank system.

The water samples were extracted in the laboratory of the University of Zambia. Approximately 500 mL of the sample was first filtered through a 47 mm GF/D (2.7 µm) glass microfiber filter and then through a GF/F (0.7 µm) glass microfiber filter into a clean vacuum flask. Some of the surface and wastewater samples had to be pre-filtered through a normal filter paper because of clogging. 200 mL of the filtered water sample was then poured into two clean 200 mL conical flasks. Using an automatic pipette, 40 µL of the 10 ppm internal standard mixture consisting of isotopically labeled pharmaceuticals (Chapter 3.2) was added to each flask and shaken well to mix.

Picture 11. Solid phase extraction method was used for extraction and concentration of the water and urine samples.

The Oasis hydrophilic-lipophilic balance (HLB) SPE cartridges were prepared by conditioning them with 3 mL of analytical grade methanol, followed by 3 mL of ultrapure water added with a 5 mL syringe. Groundwater samples used 3 cc/60 mg cartridges while surface and wastewater samples used 6 cc/200 mg cartridges. After conditioning, the cartridges were loaded with the filtered sample, not letting them dry in between. After loading the cartridges were allowed to dry in vacuum for 10 minutes. A suction pump (Chemat Technology Inc., Los Angeles, USA) was used in filtration and extraction.

3.4 Sample analysis with LC-MS/MS

The extracted samples were first stored in Lusaka in a sealed bag, protected from light and shipped to the University of Jyväskylä for identification and quantification analysis of the pharmaceutical concentrations by liquid chromatography-tandem mass spectrometry (LC-MS/MS), which was carried out in December 2016. The analysis was conducted based on the methods previously described by Ngumba et al. (2016).

The pharmaceuticals were analyzed by Waters Quattro™ Micro LC-MS/MS system. Liquid chromatography was performed on a Waters Alliance 2795 (Milford, MA, USA) system consisting of a tertiary pump, a vacuum degasser, an autosampler and a column oven (30 °C). The compounds were separated by a reverse phase column C 18 (Waters XBridgeTM 3.5 µm, 2.1×100 mm with 3.5 µm, 2.1×10 mm guard column). The mobile phase consisted of ultrapure water (A) and acetonitrile (B), both containing 0.1% (v/v) formic acid. The flow rate was 0.25 mL/min and the injection volume was 10 µL. The B gradient was maintained at 20% for the first 2 minutes, after which it was linearly increased to 100% at 3 minutes. Then, B was lowered by 20% when 5 minutes had passed and this was maintained for 2 minutes. The column was then equilibrated for 7 minutes before the next injection. The total time for each injection was 19 min.

The Micromass Quattro electron beam triangle quadrupole mass spectrometer (Micromass, Manchester, UK) was used as a detector in the tandem mass spectrometry. Nitrogen was used as the desolvation gas (500 L/h) and as the cone gas (50 L/h). The desolvation temperature and the source temperature were 200 °C and 100 °C. Argon was used as a collision gas at 2.8×10^{-4} mBar collision pressure. Mass spectrometric analysis was performed in a positive electron beam ionization mode (ESI+) and the mass spectrometer was operated in multiple reaction monitoring (MRM) with a dwell time of 200 ms and a similar delay between the channels.

3.5 Observation and interviews

Apart from the analyzed water and urine samples, additional data for this study was collected by observation and an interview. Because the data comprised by these methods was not sufficient for analysis, the data has been collected in Chapter 5.1 in the Discussion part of this thesis.

The observation was done by walking around in the study area during the field work months and taking notes on the activity related to water, wells, toilets and peri-urban agriculture. Observation was also made during sample collection, especially when talking with the local residents and when interviewing them in order to collect sample background data. Field notes were written down on a daily basis. The interview was conducted with the personnel of the closest local health facility, Chunga health clinic. The discussed topics included the perceived risks of pit latrines, the usage of dry toilets, the fate of the pharmaceuticals in the environment and the local HIV situation and antiretroviral therapy in Chunga and Madimba. The interview provided more information about the knowledge base of the health clinic personnel than about the actual situation and therefore the answers were left unanalyzed.

4 RESULTS

In this study, the occurrence of seven antibiotics (AMO, CIP, DOX, NOR, SMX, TET, TMP) and three antiretroviral drugs (3TC, NVP, ZDV) (Chapters 2.3 and 2.4) were studied in groundwater, wastewater, surface water and source-separated urine in Madimba, a peri-urban settlement of Lusaka, Zambia. The concentrations varied considerably between the different sample types, being occasionally present in groundwater, intensively present in surface and wastewater with highest reported concentrations in wastewater, and continuously present up to varying, also very high, concentrations in source-separated urine.

4.1 Measured pharmaceutical concentrations in groundwater

In groundwater, the phamaceutical contaminants were occasionally present in the groundwater samples collected from shallow wells and borehole wells. Half of the studied pharmaceuticals were detected: four out of seven antibiotics and one out of three antiretroviral drugs (Table 2). The compounds were detected in the ng/L range and the concentration of the antibiotics ranged from unquantifiable (<LOQ) to 880 ng/L (Figure 6). A summary of the detection frequency, range and median concentration of the studied pharmaceuticals is presented in Table 2, whereas Table 3 presents the mean pharmaceutical concentrations in all 26 groundwater samples.

Table 2. The detection frequencies and concentrations of the selected antibiotics and antiretroviral drugs in the groundwater samples in ng/L.

Compound	Detection frequency (%)	Concentration range	Median concentration
AMO	11.5	nd–880	760
CIP	19.2	nd–150	90
DOX	0	nd	nd
NOR	0	nd	nd
SMX	42.3	nd–660	100
TET	0	nd	nd
TMP	34.6	nd–140	60
3TC	0	nd	nd
NVP	38.5	nd–410	150
ZDV	0	nd	nd

nd: not detected

The antibiotic sulfamethoxazole was present at the highest detection frequency of 42.3% and a concentration range fluctuating from not detected to 660 ng/L (Figure 7). The frequent detection of SMX can be related to the high mobility of the compound in soil and to its other properties such as low biodegradability (Deng et al. 2016). Trimethoprim was the second most common pharmaceutical with a detection frequency of 34.6% and concentrations up to 140 ng/L, which was lower than SMX. TMP is at its optimum sorption range in the acidic soils of the study area (pH approximately 4-5.5), which together with its high distribution coefficient (K_d) makes the compound largely immobile (Chabala et al. 2014). In addition, TMP is generally administered together with sulphonamides such as SMX in a ratio of 1:5, which further explains its lesser concentrations due to its lower mass load into the environment (WHO 2016).

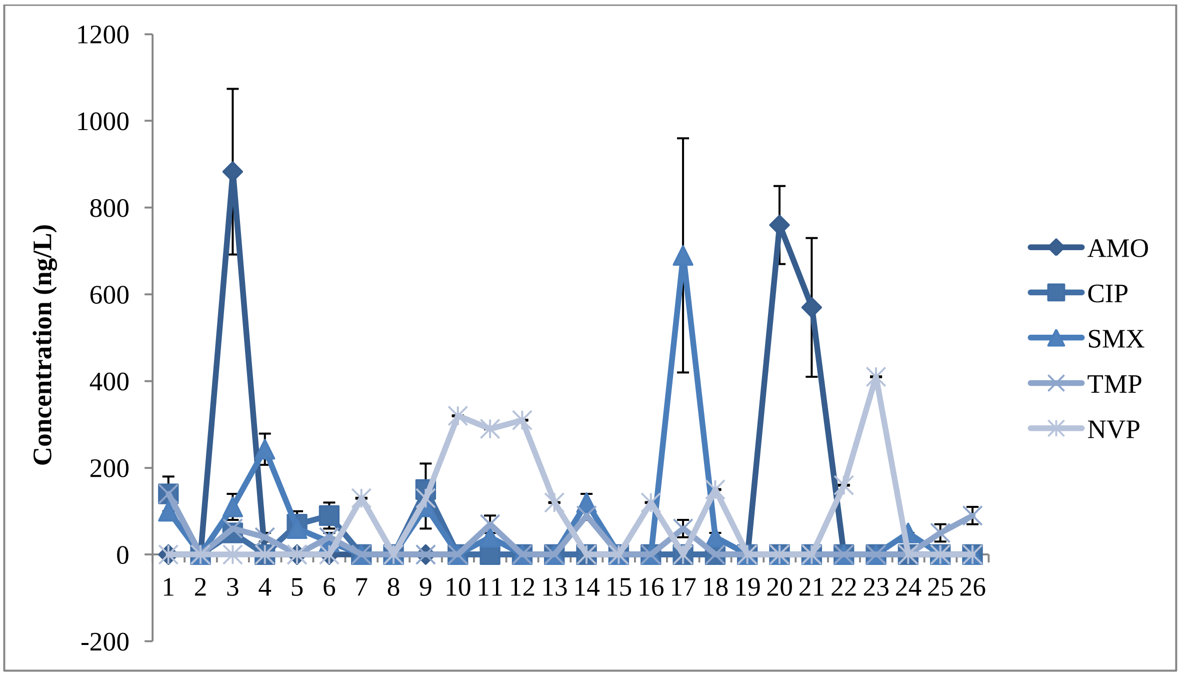


Figure 7. AMO, CIP, SMX, TMP and NVP were detected in the ng/L range in the 26 groundwater samples of Lusaka.

CIP was the only compound in the pharmaceutical class of fluoroquinolones that was detected in some well waters. The maximum concentration was 150 ng/L and the detection frequency was 19.2%. The lower detection frequency was attributed to the high K_d of fluoroquinolones in soil (Le-Minh et al. 2010). CIP is also a less consumed antibiotic because of its more expensive price (Ngumba 2017 - unpublished). Amoxicillin was present in only 11.5% of the wells with a maximum concentration of 880 ng/L, yet it is one of the most prescribed antibiotics in the world and expected to be highly mobile in soil (Kim et al. 2012). The concentrations of AMO that were detected in the few wells are attributed to contamination of water originating from the pit latrines in close proximity to the wells. Tetracycline and doxycycline were not detected in the groundwater because both compounds have high distribution coefficients in soils and are largely considered immobile (Kim et al. 2012). However, relatively high concentrations of these were found in the surface water and wastewater samples.

Nevirapine was the only antiretroviral drug that was detected in the groundwater samples with a maximum concentration 410 ng/L and a detection frequency of 38.5%. This is most likely linked to the mobility to groundwater as well as to non-biodegradability of NVP, which leads to persistence in the environment (Jain et al. 2013). Lamivudine and zidovudine were hardly detected in groundwater despite their high concentrations in wastewater, surface water and the source-separated urine. This can be attributed to the acidic soils of the study area (Chabala et al. 2014) combined with the high adsorption capacities of 3TC and ZDV, which binds them to the soil particles.

Table 3. Concentrations of the selected pharmaceuticals in the groundwater samples in ng/L.

	AMO	CIP	DOX	NOR	SMX	TET	TMP	3TC	NVP	ZDV
GW1	-	140 (40)*	-	-	100 (20)	-	140 (20)	-	-	-
GW2	-	-	-	-	-	-	-	-	-	-
GW3	883 (191)	50 (10)	-	-	110 (30)	-	60 (10)	-	-	-
GW4	-	-	-	-	243 (36)	-	40 (10)	-	-	-
GW5	-	70 (30)	-	-	60 (10)	-	-	-	-	-
GW6	-	90 (30)	-	-	29 (10)	-	40 (10)	-	-	-
GW7	-	-	-	-	-	-	-	-	130 (10)	-
GW8	-	-	-	-	-	-	-	-	-	-
GW9	-	150 (60)	-	-	110 (50)	-	-	-	130 (40)	-
GW10	-	-	-	-	-	-	-	-	320 (90)	-
GW11	-	-	-	-	40 (10)	-	70 (20)	-	290 (20)	-

GW12	-	-	-	-	-	-	-	-	310 (10)	-
GW13	-	-	-	-	-	-	-	-	120 (10)	-
GW14	-	-	-	-	120 (20)	-	90 (10)	-	-	-
GW15	-	-	-	-	-	-	-	-	-	-
GW16	-	-	-	-	-	-	-	-	120 (20)	-
GW17	-	-	-	-	690 (270)	-	60 (20)	-	-	-
GW18	-	-	-	-	40 (10)	-	-	-	150 (10)	-
GW19	-	-	-	-	-	-	-	-	-	-
GW20	760 (90)	-	-	-	-	-	-	-	-	-
GW21	570 (160)	-	-	-	-	-	-	-	-	-
GW22	-	-	-	-	-	-	-	-	160 (10)	-
GW23	-	-	-	-	-	-	-	-	410 (30)	-
GW24	-	-	-	-	50 (0)	-	-	-	-	-
GW25	-	-	-	-	-	-	50 (20)	-	-	-
GW26	-	-	-	-	-	-	90 (20)	-	-	-

*Mean (±SD); - not detected

Pharmaceutical residues were detected in 22 out of 26 groundwater wells. Contaminants were present in all five borehole wells, what refers to a broader contamination of the vulnerable aquifers of Lusaka. Interestingly, all the four shallow wells without detected pharmaceutical contamination (levels below limit of quantification) were estimated to be especially close to adjacent pit latrines, between 5 and 10 meters. Whether the shallow well was protected or not did not affect the detected contamination. In addition, at least half of the uncontaminated shallow wells were situated in the water-logged parts of the study area. Based on the limited data of this study it cannot be concluded that closer proximity to pit latrines or higher groundwater table would directly increase the levels of pharmaceutical contamination in the groundwater, however it is apparent that the high density of pit latrines and the high disease burden together with corresponding consumption of medicines have contributed to the pharmaceutical contamination indirectly.

4.2 Measured pharmaceutical concentrations in surface water and wastewater

The pharmaceutical residues were detected in considerably higher levels in the surface and wastewater samples than in the groundwater samples. All the studied pharmaceuticals were detected in the surface and wastewaters of the study area with concentrations ranging from ng/L to µg/L levels (Table 4). The concentrations for most of the compounds were also significantly higher than the concentrations reported in previous studies in other locations (see Chapter 5.1).

Table 4. Concentrations (ng/L) of the selected pharmaceuticals in the surface water samples from Chunga River and in the wastewater samples from Matero wastewater stabilization ponds and Chunga WWTP effluent.

	Surface water		Wastewater		
	Chunga River A	Chunga River B (WWTP)	WW pond A influent	WW pond A effluent	WW pond B
AMO	2500 (660)*	3410 (440)	3270 (690)	5580 (1880)	4460 (570)
CIP	400 (90)	540 (70)	740 (80)	230 (30)	170 (10)
DOX	2730 (610)	3260 (590)	4490 (810)	5280 (1190)	4980 (70)
NOR	nd	nd	100 (20)	80 (20)	nd
SMX	11800 (1200)	7810 (740)	33300 (1890)	30040 (3420)	41080 (2870)
TET	2200 (700)	4220 (740)	220 (20)	4590 (540)	nd
TMP	2410 (20)	510 (50)	32670 (1570)	1770 (160)	360 (60)
3TC	49700 (4000)	42630 (3660)	118970 (9450)	55760 (5480)	232920 (17350)
NVP	210 (30)	220 (30)	680 (60)	1720 (250)	1870 (90)
ZDV	1280 (400)	9670 (1290)	66590 (4650)	37140 (2560)	nd

*Mean (±SD); WW: wastewater; nd: not detected

The concentrations of antibiotic residues in the surface water samples ranged from undetectable amounts (<LOQ) up to 11800 ng/L. In surface waters, SMX was detected at the highest concentration of 11800±1200 ng/L at the Chunga River sampling site A (Figure 8). NOR was not detected from either of the two surface water sampling sites while the concentrations of the other antibiotics ranged between these two values. Correspondingly, the concentration of individual antibiotics in the wastewater of the Matero wastewater stabilization ponds fluctuated between 100 and 33300 ng/L. The concentrations in WWTP effluent ranged from 80 to 30040 ng/L. It was not possible to take a sample from the WWTP influent. SMX was the most dominant antibiotic in both the influent and effluent waters with mean concentrations of 33300±1890 ng/L and 30040± 3420 ng/L, respectively (Figure 8).

The concentrations of antiretroviral drug residues in the surface water samples ranged from <LOQ up to 49700 ng/L. 3TC was detected at the highest concentration of 49700±4000 ng/L followed by ZDV with a lesser concentration of 9670±1290 ng/L (Figure 8). NVP was detected at the lowest concentrations of 220±30 ng/L (Figure 9). The large variation in the concentration can be attributed to the different excretion rates of the antiretroviral drugs in the unchanged form, discussed in Chapter 3.2. The three antiretroviral drugs constitute the first line daily dose antiretroviral regimen for people living with HIV so their usage levels should not differ much. The amount of antiretrovirals was high also in the wastewater with maximum concentration of 232920 ng/L. The wastewater stabilization pond influent concentrations were ranging from 680 to 118970 ng/L and from 1720 to 55760 ng/L in the stabilization pond effluent, respectively. 3TC was the most dominant antiretroviral residue in the effluent with a mean concentration of 55760±5480 ng/L followed by ZDV with mean concentrations of 37140±2560 ng/L (Figure 8). NVP was present in the lowest amounts with a mean concentration of 1720±250 ng/L (Figure 9).

At the Chunga River sampling site A, the concentrations of three pharmaceuticals (SMX, TMP and 3TC) were higher than the levels at the Chunga River WWTP effluent discharge site B, which is the result of direct domestic wastewater discharges into the river. The notably high concentration of antibiotics and antiretroviral drugs in the surface and wastewater is directly related to the high prevalence of diseases in the study area, especially HIV/AIDS that require vast usage of both types of medicines as part of the antiretroviral therapy. Also inadequate sanitation measures and inefficient wastewater treatment contribute to high levels of pharmaceuticals in the wastewater, which is often illegally discharged untreated to the surface water bodies such as the Chunga River. The measured high concentration of pharmaceuticals can therefore be attributed to direct discharges of untreated domestic wastewater from the adjacent low-income housing areas, but also discharges from the Chunga WWTP and Matero wastewater stabilization ponds.

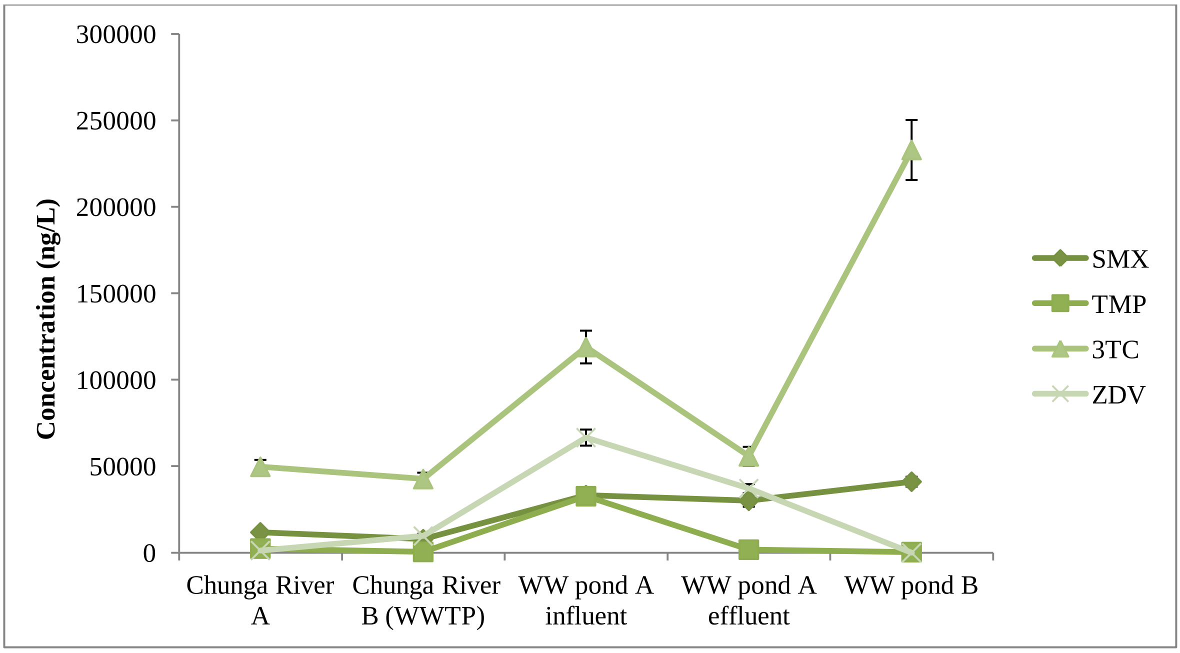


Figure 8. SMX, TMP, 3TC and ZDV were detected at the overall highest concentrations in the surface and wastewater samples of Lusaka.

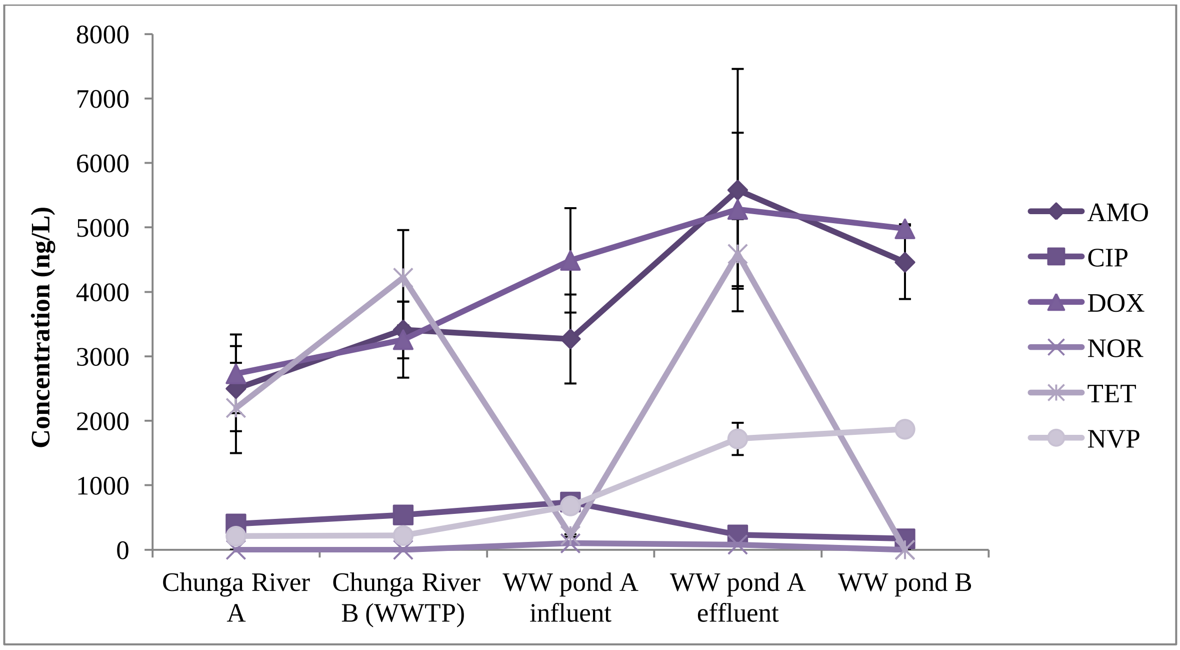


Figure 9. AMO, CIP, DOX, NOR, TET and NVP were present at relatively lower concentrations in the surface and wastewater samples of Lusaka.

4.3 Measured pharmaceutical concentrations in source-separated urine

The concentration of the target pharmaceuticals in the studied source-separated urine were several orders of magnitude higher than in the wastewater. The concentrations ranged from below µg/L up to mg/L level, which is why the concentrations have been presented in µg/L instead of ng/L. All the studied source-separated urine samples contained at least four different pharmaceutical compounds. Trimethoprim, lamivudine and sulfamethoxazole had the overall highest measured concentrations at 12800 µg/L, 10010 µg/L and 7740 µg/L (Figure 10), while TMP, DOX and 3TC were detected in all of the urine samples in different concentrations (Table 4).

Table 4. Concentration of antibiotics and antiretroviral drugs in the source-separated urine samples in µg/L.

	AMO	CIP	DOX	NOR	SMX	TET	TMP	3TC	NVP	ZDV
U1	82.1 (9.3)*	5.5 (0.5)	14.0 (1.5)	-	-	-	2.4 (0.3)	224.8 (25.9)	-	-
U2	13.5 (6.4)	3.0 (1.0)	17.0 (1.0)	0.2 (0.0)	-	-	6.1 (1.6)	3.3 (0.7)	-	-
U3	30.8 (5.4)	8.7 (1.4)	13.7 (1.1)	5.3 (1.8)	2763.3 (341.3)	0.7 (0.1)	7599.9 (427.5)	2.0 (0.5)	-	-
U4	-	5.2 (1.5)	19.6 (2.0)	-	-	0.9 (0.2)	4.4 (0.6)	3.6 (0.6)	-	-
U5	10.6 (1.2)	662.5 (130.9)	16.2 (1.3)	3.2 (1.8)	-	2.8 (0.6)	5.4 (0.8)	35.5 (2.8)	5.0 (0.2)	-
U6	-	3.2 (0.3)	2.0 (0.7)	2.4 (0.8)	4.5 (1.1)	-	0.7 (0.054)	1.9 (0.2)	-	-
U7	9.3 (4.2)	3.9 (1.3)	13.0 (1.4)	4.0 (0.5)	7743.4 (566.6)	2.8 (0.6)	12796.7 (368.5)	10013.7 (806.1)	-	-
U8	-	-	4.3 (0.898)	1.2 (0.3)	-	-	1.7 (0.4)	1.9 (0.1)	-	-
U9	12.5 (2.6)	3.1 (0.4)	18.8 (1.1)	3.4 (0.5)	-	-	4.1 (0.2)	5131.6 (166.7)	-	-
U10	306.2 (63.6)	7.81 (1.1)	12.3 (1.8)	1.9 (0.7)	1662.4 (357.9)	-	1571.6 (55.1)	1249.0 (87.8)	-	-

*Mean (±SD); - not detected

All the studied antibiotics were detected in the source-separated urine samples with frequencies between 40–100% (Table 5). Trimethoprim and doxycycline were detected in all the studied samples while tetracycline and sulfamethoxazole were found in four out of 10 samples. TMP had the highest measured antibiotic concentration at 12800 µg/L, followed by SMX (7740 µg/L), CIP (660 µg/L), AMO (310 µg/L), DOX (20 µg/L), NOR (5.3 µg/L) and TET (2.8 µg/L). TMP was also largely present in the groundwater as well as surface and wastewater samples, which makes it a widely spread contaminant in the study area. The particularly high concentrations of TMP and SMX can be explained by their regular use for treatment of various opportunistic microbial infections, often as a combination of the two as co-trimoxazole (WHO 2016).

Table 5. The detection frequencies and concentrations of the selected pharmaceuticals in the source-separated urine in µg/L.

Compound	Detection frequency (%)	Concentration range	Median concentration	Mean concentration
AMO	80	nd–310	13	58.1
CIP	90	nd–660	5.2	78.1
DOX	100	2–20	13.8	13.1
NOR	80	nd–5.3	2.8	2.6
SMX	40	nd–7740	1662.4	2434.7
TET	40	nd–2.8	0.9	1.4
TMP	100	0.7–12800	4.9	2199
3TC	100	1.9–10010	19.6	1667.8
NVP	10	nd–5	5	5
ZDV	0	nd–nd	nd	nd

nd: not detected

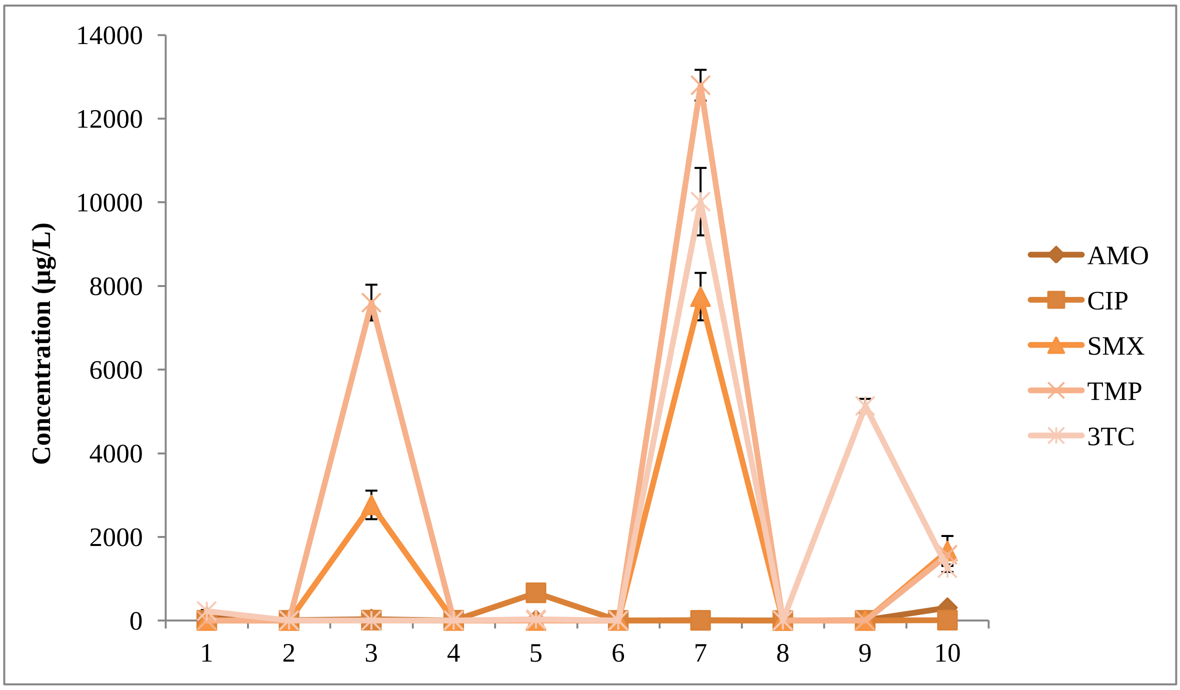


Figure 10. AMO, CIP, SMX, TMP and 3TC were detected at the overall highest concentrations in mg/L range in the 10 source-separated urine samples collected in Lusaka.

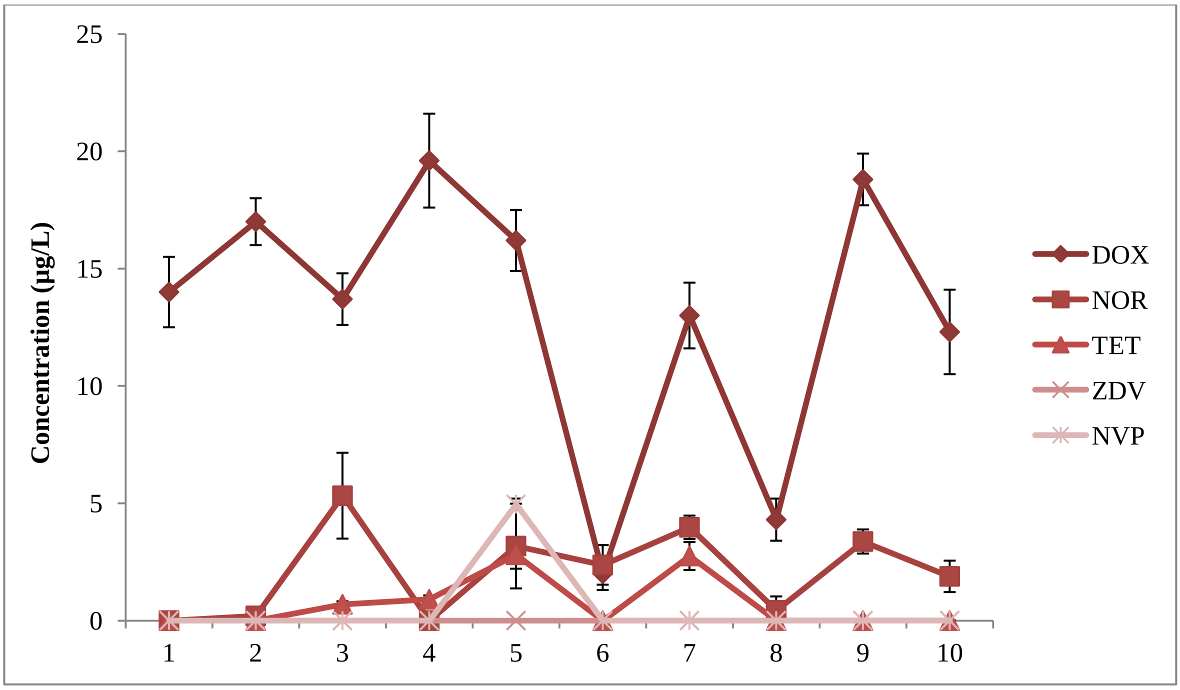


Figure 11. DOX, NOR, TET, ZDV and NVP were detected in relatively lower concentrations in the 10 source-separated urine samples collected in Lusaka.

The occurrence of antiretroviral drugs in the source-separated urine varied significantly between the samples. 3TC was detected in all of the samples with also the highest frequency (100%) and concentration (10010 µg/L) (Figure 10.) while NVP was detected in only one sample and ZDV concentrations were below the limit of quantification (Figure 11). 3TC was also present in high amounts in the surface and wastewater samples, but not in any of the groundwater samples. The variation in the concentrations of the three antiretroviral drugs in urine can be attributed to the levels of consumption, but also to the different excretion rates of the compounds in the unchanged form as discussed earlier.

In the light of these results, it is evident that the pit latrines used widely in the study area are contributing to the contamination of the adjacent shallow groundwater wells not only through pathogens, but also through pharmaceutical substances. The pharmaceutical concentrations in the Chunga River where wastewater is being discharged by households and the wastewater treatment plant, as well as in the wastewater stabilization ponds, are especially elevated. The irrigation of food crops with water from such surface and wastewater sources can be considered unsafe as it has been detected that the pharmaceuticals can be incorporated by the irrigated plants (Eggen et al. 2011; Azanu et al. 2016).

Careful consideration is also required when evaluating whether source-separated urine containing high concentrations of antibiotic and antiretroviral drug residues should be used as a fertilizer, especially in case of food crops. In areas with high disease prevalence and consumption of pharmaceuticals such as the peri-urban areas of Lusaka, it cannot necessarily be recommended. This issue will be further elaborated in Chapter 5. together with suggested preventive measures.

5 DISCUSSION

5.1 Field observation

Some field observation notes were accrued during the data collection for this research regarding the water and sanitation situation, usage of the built UDDTs as well as the viability of the practical arrangements of the sanitation and urine fertilization system in the peri-urban area of Madimba, Lusaka. As this thesis was done in collaboration with the Innovative Sanitation for Peri Urban Areas in Lusaka project presented in Chapter 2.1.3, the collected notes were included in this work. As the data was insufficient for analysis, the observations are presented here.

Regarding the water supply situation in the peri-urban communities such as Madimba, the daily water economy is often a mixture of tap water and groundwater. A household can fetch tap water for drinking from a water kiosk while using groundwater from a shallow well for other activities such as cleaning and washing. It also became evident that there are private, shared and semi-public shallow wells in the study area. The semi-public wells were described as owned and managed by one of the households but located centrally enough for several dozens of people to use them. Many of the shallow wells are unprotected, but some protected wells also exist.

During sample collection and observation it was noticed that some of the Matero wastewater stabilization ponds were suffering from eutrophication and overgrown while others were nearly empty. Nevertheless, the wastewater from the ponds was used for irrigation of vegetable gardens grown next to the stabilization ponds. Also the discharged WWTP effluent and the contaminated river waters were directly used for irrigation of food crops in the area. Agricultural plots have been established even inside the area of the WWTP facility.

What comes to sanitation, it became evident that several households with urine-diverting dry toilets have started to use the dry toilets in the same manner as pit latrines after encountering problems either with the toilet structures, such as clogging urine-diversion pipes, or possible challenges linked to ownership or cultural issues. In any case, several dry toilet owners were not separating the urine from other toilet waste and many of those who were, did not utilize the urine. Instead, it was often emptied from the full container by pouring the urine onto the ground, for instance behind the toilet or in a garbage pit. As the urine was analyzed for high contents of pharmaceuticals, the measure spreads these micropollutants and nutrients contained by the urine directly into the environment. Also most of Madimba is characterized by a high groundwater table, meaning that water can easily act as a conveyor of pollutants.

The sanitation needs and infrastructure in the study area do not match the demand for local agricultural fertilizers or the infrastructure that would be essential in order to use the source-separated urine as a fertilizer. For instance, an average Zambian household of five people produces approximately 10 liters of urine per day, while the containers used in the urine-separating toilets of Madimba hold a volume of 20 liters. This would mean changing the container every few days and storing several containers of urine for over 6 months if the hygienization recommendations given by WHO (2006) would be followed. Currently infrastructure for storage of such volumes of source-separated urine does not exist. However, only 20% of the residents of the study area have backyard gardens (GDTF 2017a), what indicates an excessive production of fertilizer compared to the local need and capacity to utilize it in peri-urban agriculture. Also the measured high pharmaceutical concentrations in the source-separated urine pose an impediment to the direct agricultural use of the urine collected in Madimba. It may have been a blessing in disguise that the collection of urine has been disturbed in many households having UDDTs.

A potential solution to the disposal measures and the mismatch of the potential supply and demand capacity could be arranging safe collection and transport of the source-separated urine. It could then be either disposed of in a safe manner or processed into contaminant-free agricultural fertilizers by different available treatment options such as struvite precipitation, evaporation, electrodialysis, electrolysis and combination of nitrification and distillation as presented in Chapter 2.3. The fertilizer products could be used in the backyard gardens and the excess sold and transported for example to the close-by agricultural areas. This measure would also have the potential of local sustainable business development. Alternatively, the source-separated urine could be used for fertilization of energy plants. In that case, it would be important to prevent environmental contamination with the micropollutants present in the urine and evaluate the antimicrobial resistance risks deriving from the exposure of soil microbes to the pharmaceutical substances.

5.2 Contamination levels in comparison to other locations

The pharmaceutical concentrations detected in Lusaka, Zambia were overall higher than the levels reported in previous studies. While groundwater and surface water contamination was on the same or slightly elevated level compared to other locations in Africa, concentrations found in wastewater were significantly higher than elsewhere. When compared to studies conducted in industrialized countries, all concentrations measured in Lusaka were on high levels. In the developing world, concentrations of the same magnitude and similar contamination patterns have been detected for instance in Kenya (K'oreje et al. 2016; Ngumba et al. 2016) and in other locations in sub-Saharan Africa. In the light of these studies it has become evident that the ubiquitous pharmaceutical contamination is even more serious challenge in this region and in the global South than it is in the global North.

In the groundwater of Lusaka, the concentrations were generally in the same level or modestly higher than elsewhere. The concentrations of antibiotics detected in this study in the groundwater of Lusaka were higher than reported in previous studies in different locations. However, the results were within one order of magnitude with the previous studies. For instance K'oreje et al. (2016) have measured maximum SMX concentrations of 30 ng/L in Nairobi, Kenya, while in the USA, Schaider et al. (2014) have reported concentrations of up to 113 ng/L in Barnstable County of Massachusetts and Fram & Belitz (2011) have measured SMX concentrations of up to 170 ng/L in California. The maximum SMX concentration of 690 ng/L detected in Lusaka vastly exceeds these results, while concentrations of AMO were even higher (883 ng/L). On the other hand, Fick et al. (2009) have reported considerably higher concentrations (14000 ng/L) of CIP in groundwater in India compared to this study, but their sampling location was close to a pharmaceutical production factory which explains the extremely high results. TMP has been detected in the drinking water supply of California, USA at concentrations up to 18 ng/L, which were lower than detected in Lusaka (up to 140 ng/L) (Fram & Belitz 2011).

What comes to antiretroviral drug traces in groundwater, limited studies exist. K'oreje et al. (2016) have detected nevirapine in shallow well waters in the Kenyan cities of Nairobi and Kisumu with highest concentrations measured in a densely populated informal settlement. The compounds were present in higher concentrations than those measured in this study (1600 ng/L compared to 410 ng/L). K'oreje et al. had also similar findings on the environmental persistence of NVP as found in this study: the environmental fate of nevirapine seems to be different than of the other antiretroviral drugs lamivudine and zidovudine, as it is more mobile to groundwater. The higher log K_{ow} value of NVP (2.5 versus -1.4/0.05) supports this finding. Accordingly, much higher concentrations of ZDV were measured in the surface water than in the groundwater in Kenya. In Lusaka, 3TC and ZDV were hardly detected in groundwater despite their high concentrations in surface and wastewater as well as in the source-separated urine, which implies that these two compounds are not largely mobile to groundwater.

In the wastewater of Lusaka, the measured concentrations were remarkably higher in most of the studied pharmaceutical compounds than what has been reported elsewhere in the world in previous studies. For example, one of the thus-far highest measured concentrations for AMO was found to be 120 ng/L measured in WWTP effluent in Italy by Andreozzi et al. (2004) compared to 5580 ng/L in Lusaka. CIP has been detected at higher concentrations than in this study, up to 5600 ng/L in WWTP effluent in the USA as compared 740 ng/L in Lusaka (Batt et al. 2006). DOX concentration has been detected at 46 ng/L (compared to 5280 ng/L in Lusaka) and NOR concentration 112 ng/L in the WWTP final effluent in Canada (Miao et al. 2004), which is also slightly higher than the values measured in this study (100 ng/L). SMX has been detected at lower concentrations up to 6000 ng/L in WWTP effluent in the USA (Batt et al. 2006) and 1310 ng/L in WWTP in Bolivia (Archundia et al. 2017), compared to 41080 ng/L in Zambia. The maximum concentration of TET has been 977 ng/L in WWTP effluent in Canada and TMP concentration has been 336 ng/L in WWTP effluent in Bolivia (Miao et al. 2004; Archundia et al. 2017), compared to 4590 ng/L and 32670 ng/L in Zambia. The maximum reported concentrations for antiretroviral drugs in wastewater have been 31070 ng/L for 3TC, 2080 ng/L for NVP in Kenya and 564 ng/L for ZDV in effluent wastewater in Germany (K’oreje et al. 2016; Prasse et al. 2010). The corresponding values in Zambia were 232930 ng/L, 1870 ng/L and 66590 ng/L, meaning that while NVP has been detected in slightly higher concentration in Kenya, the other two concentrations measured in Zambia were 10-30 times higher.

The concentration of antibiotics and antiretroviral drugs in the surface water were on a similar level than data from other sub-Saharan countries of South Africa, Kenya, Mozambique and Ghana. These results were within one order of magnitude relative to this study. For example, Segura et al. (2015) have reported antibiotic concentrations in these four countries, with SMX concentrations ranging from below the limit of quantification (<LOQ) to 9640 ng/L in Ghana, from <LOQ to 49562 ng/L in Kenya, from 511 to 53828 ng/L in Mozambique and from 3.3 to 10568 ng/L in South Africa, compared to 7810 to 11800 ng/L in Zambia. Maximum TMP concentrations were measured at 1374 ng/L in Ghana, 11383 ng/L in Kenya, 6223 ng/L in Mozambique and 5875 ng/L in South Africa, compared to 2410 ng/L in Zambia. For TET, maximum detected concentrations were 465 ng/L in Ghana and 434 ng/L in Kenya while not detected in Mozambique or South Africa. In addition, DOX was occasionally present in low quantities. Agunbiade & Moodley (2014) on the other hand have detected TET in South African surface waters, ranging from 640 to 5680 ng/L, compared to 2200 to 4220 ng/L in Zambia. They also reported concentrations of CIP ranging from 710 to 16900 ng/L as compared to 400 to 540 ng/L in Zambia and SMX with a median concentration of 3680 ng/L as compared to the range of 7810 to 11800 ng/L in Zambia. In resource-limited settings, low-cost antibiotics such as SMX and TMP are generally detected more frequently and in higher concentrations than more expensive drugs (Segura et al. 2015).

Antiretroviral drugs have been detected in the African surface waters as well: 3TC has been measured at concentrations ranging from <LOQ to 167100 ng/L and ZDV from <LOQ to 17410 ng/L in Kenya (K'oreje et al. 2016). These findings are on the same level of magnitude with this study as compared with the concentrations of 42630 to 49700 ng/L for 3TC and 1280 to 9670 ng/L for ZDV. In addition, NVP has been detected ranging from <LOQ to 177 ng/L in South Africa and from 30 to 5620 ng/L in Kenya, compared to 210 to 220 ng/L in Zambia (Wood et al. 2015; K'oreje et al. 2016). Also Ngumba et al. (2016) have measured high concentrations of these pharmaceuticals in Kenya. When comparing these results to concentrations measured in industrialized countries, the concentrations in Zambia and other African countries are significantly higher exceeding the levels in industrialized countries by several orders of magnitude. For instance, ZDV concentrations up to 170 ng/L have been detected in German river waters while 3TC was not detected at all (Prasse et al. 2010), compared to 9670 ng/L and 49700 ng/L measured in the Chunga stream in Zambia.

Segura et al. (2015) have found a significant difference in antibiotic concentrations in surface water between high-income and low- and lower-middle income countries, yet not between high and upper-middle income countries. A relationship exists between the occurrence of pharmaceuticals in the environment and the income inequality between these countries, because the wastewater collection and treatment is often inadequately arranged in the both low- and lower-middle income countries. With wastewater being the most important source of pharmaceuticals in the environment, the effects to aquatic life are expected to be more pronounced in countries with inadequate and unsafe sanitation infrastructures. Furthermore, the residents of low- and lower-middle income countries are more vulnerable to infectious diseases requiring medication and cheap over-the-counter medicines are also more available in lower-income countries. The existing knowledge on pharmaceutical contamination suggests that the situation is especially worrying in the African continent. The concentrations measured in this study and in other recent studies conducted in sub-Saharan Africa indicate a serious wide-scale contamination of urban hydrological cycles with antibiotics and antiretroviral drugs in the entire region.

What comes to source-separated urine, Bischel et al. (2015) recently studied pharmaceutical concentrations in source-separated urine in eThekwini, South Africa. Maximum concentrations of 6800 µg/L for SMX and 1300 µg/L for TMP were measured. These results are lower compared to those found in this study, yet in the same order of magnitude: in Zambia, SMX was detected at a maximum concentration of 7740 µg/L and TMP at 12800 µg/L. These pharmaceuticals do not degrade to a large extent during storage of the source-separated urine, even in the 6 month storage period recommended by WHO (2006) for hygienization of the urine (Bischel et al. 2016; Jaatinen et al. 2016). Data is limited, but based on this study and the results of Bischel et al. (2016) it looks like the source-separated urine is likely to contain high volumes of biologically active yet non-biodegradable and potentially hazardous pharmaceuticals in HIV-prevalent areas such as in sub-Saharan Africa.

5.3 Recommendations for management of pharmaceutical contaminants

Environmental issues and human health often go hand in hand, and in the peri-urban settlement of Madimba as well as in similar settings with high disease prevalence and consequent pharmaceutical consumption, the use of source-separated urine as an agricultural fertilizer may pose considerable ecological, human health and socio-economical risks. The high concentrations of pharmaceuticals detected in source-separated urine analyzed in this study call for careful considerations to be made when urine is intended for direct use as a fertilizer in the production of food crops. Also when used in non-food crop agriculture, protective measures should be taken in order to avoid environmental contamination.

Perhaps the most important socio-economical and health risk linked to the pharmaceutical residues in the environment and through the uptake of food crops is the antimicrobial resistance in the pathogens, which can lead to development of resistant microbial populations. Recent studies have revealed that pharmaceutical contamination in the hydrological cycles have the potential of developing antimicrobial resistance through antibiotic resistance genes (ARGs) that have been detected especially in wastewaters and disseminated in the environment (Segura et al. 2015; Archundia et al. 2017). The continuing exposure to low doses of pharmaceuticals can lead to development of antimicrobial resistant bacteria: for instance soil bacteria could generate resistance through low but constant concentrations in groundwater (Archundia et al. 2017). ARGs have also been detected in all source-separated urine samples that were studied in South Africa by Bischel et al. (2015).

For instance Eggen et al. (2011) and Azanu et al. (2016) have found out that significant amounts of pharmaceutical residues can be incorporated by the food crops when the crops are fertilized or irrigated with materials containing these pollutants. According to WHO (2017a), resistance to pharmaceuticals used in the treatment of HIV/AIDS has been increasing, and the matter requires urgent attention. Especially in areas with high disease prevalence and high environmental concentrations of pharmaceuticals, the risk of antimicrobial resistance development is acute and serious and it might necessitate the use of more expensive second-line drugs in the future (ibid). This is problematic since majority of the people receiving HIV treatment live in the low- or middle-income countries, and cannot necessarily afford these new drugs. Most of these households are already poor, and the situation would be likely to get even worse with the increasing development of antimicrobial resistance in the HI-virus.

As data on the consumption of medicines, especially in the case of often sensitive HIV medications can be non-reliable, it would be suggested to test the pharmaceutical residue levels before designating the source-separated urine for fertilizer use. If the levels are found to be elevated and/or constant, the urine should be treated for removal of pharmaceutical residues before application. In the peri-urban settlement of Madimba, a setting with insufficient sanitation infrastructure, high groundwater table, a heavy disease burden and a social stigma attached to HIV/AIDS, it would be recommended that all urine would be source-separated and collected for further disposal or treatment. It could be the best to avoid promoting direct fertilizer use of any urine, as if only urine from households with daily medication patterns or otherwise abundant use of medicines should be collected, this could predispose such households to stigmatization and discrimination. Fear of stigma could also cause the households to not report their medication patterns honestly, as was the case during the sample collection, so dividing urine into “usable” and “unusable” volumes based solely on statements would probably be risky.

Above all, the vulnerable groundwater aquifers of Lusaka should be protected by moving from the traditional pit latrines to safer sanitation systems. Rethinking the urban and peri-urban sanitation infrastructure is needed in order to achieve safe and sustainable sanitation for the residents of the study area Madimba and other informal settlements in Zambia. The current sanitation system is inadequate and poses many potential risks, not least because of the detected concentrations of pharmaceutical residues in the peri-urban hydrological cycle and in the source-separated urine. Grönwall et al. (2010) have suggested that the water access strategies of the urban poor depend on the local hydrogeological conditions, the cultural and political situation and the strategies of water providers and city governments. According to Kawanga (2005), the integration of the peri-urban areas of Lusaka together with their ecological, socio-economic and cultural backgrounds has not been sufficiently taken into account in policy making. Furthermore, the infrastructure project designers often have training in conventional, high-technology and high-cost systems less suited to peri-urban contexts, which has played a part in the escalation of the unsatisfactory state of sanitation in the peri-urban areas such as Madimba (ibid).

First of all, an improved, safe, properly dimensioned and widely accepted sanitation system would be needed. On-site sanitation facilities such as UDDTs would be a viable option as a centralized system would be costly to implement. Secondly, these facilities should preferably separate the urine fraction from the feces fraction, as most of the nutrients and micropollutants such as pharmaceuticals are concentrated in the urine fraction. This should be collected and either disposed of in a safe manner that would not allow for spreading of the micropollutants and nutrients into the environment, or alternatively, treated in an appropriate manner for production of micropollutant and pathogen free agricultural fertilizer, depending on the desired outcome. Treatment options for removal of pharmaceutical traces in developing country contexts could be for instance electrodialysis, struvite precipitation, nitrification and solar evaporation, electrolysis and a combination of nitrification and distillation, however many of these are still on bench or pilot levels in their development (Pronk & Koné 2009; Udert et al. 2015). The approach should be market-driven: investment and operation costs of the urine collection and treatment system should be low and the added value should be taken into account. The system should also be easy to operate with locally supplied chemicals and possible to fix with locally supplied spare parts in case of technical failures and disruptions. As Zambia has recently suffered from major power cuts due to its hydroelectric power plants running on an inadequate capacity, systems that could be run without a constant power source or would incorporate an independent off-grid power source such as solar power could be the most reliable. A safe way to utilize human excreta in agriculture would be an opportunity to close the nutrient loop in the peri-urban water and sanitation management, and a step towards more comprehensive food security. Also direct fertilizer use for non-food crops such as energy crops could be an option if arranged in a controlled manner.

Based on the field observation notes, wastewater containing high concentrations of pharmaceuticals is used for irrigation of food crops in the peri-urban areas of Lusaka. The measure cannot be considered safe, yet pharmaceutical concentrations are not the only problem related to irrigation with wastewater, as high levels of heavy metal contamination have also been measured from food crops at the Chunga area in northwestern Lusaka, where this study was also taking place (Holden et al. 2009). Holden (2010) also found out that the perceived risks due to environmental contamination are not high among the Zambian policy makers or the urban farmers of Lusaka. According to the study, food safety was not perceived as an issue in Zambia, with food security and volumes of food being more important priorities. However, the results obtained during this study would call for re-examining the public perceptions of risks resulting from environmental contamination.

Further research is needed in many areas this work has touched upon. The occurrence of pharmaceutical contamination in the global South is not largely investigated, and the scarce results look rather serious especially in the region of sub-Saharan Africa. The effects of antiretroviral drugs in the environment are insufficiently studied even in the global scale. Also the relationships between pharmaceutical contamination and antimicrobial resistance would need to be recognized more thoroughly and more knowledge on antimicrobial resistance genes (ARGs) and their dissemination in the environment would be needed in order to enable a more comprehensive evaluation of the risks posed by pharmaceutical contamination of the environment. As the concentrations of pharmaceuticals in wastewater and source-separated urine were very high in Zambia, it would be extremely useful to study the occurrence of ARGs contained by them, and also the relationships between people, food crops and ARGs reaching the plants via water or urine containing such genes.

In addition, effective and appropriate technological solutions for micropollutant removal and/or nutrient recovery from source-separated urine should be further investigated and developed. These technologies should preferably be implemented in small- to medium-scale and with relatively low costs in order to be suitable for developing country contexts where environmental contamination seems to be high. Also the development of systems where non-food crops such as energy plants could be grown by utilizing the nutrient value of source-separated urine and wastewater containing micropollutants, including pharmaceuticals, in a controlled manner would be needed.

5.4 Limitations and ethical questions

There are some uncertainties arising from limitations in the sampling of this study. The sampling points were chosen by using convenience sampling as the sampling method, which may lead to bias in the research. The researcher is always using their subjective judgment when using non-random selection of research subjects. This may undermine the drawing of more general inferences about the population in question, and it has been suggested that convenience sampling should not be generalized to apply to the studied population as a whole. Furthermore, the selected subjects may not be applicable to the research problem and the researcher risks collecting poor quality data, but this should not be the case in this study (Etikan et al. 2015). However, convenience sampling is well suited for bench scale studies which aim at monitoring the need for further research.

There might be some room for error in the sampling and sample preparation phases of the study as well, arising from the limited conditions and equipment. For instance, some surface and wastewater samples were taken from a depth of only 1-2 cm as it was not possible to take them from any deeper in the water body. The samples were stored in a refrigerator that was not always on due to the daily power cuts and therefore could have gotten warmer than supposed. The samples could have contained minimal amounts of contaminants, since the sample bottles were washed with the same groundwater that was later analyzed to include pharmaceuticals. Also the laboratory equipment was washed with tap water which was not analyzed. During the sampling of the surface and wastewater samples, it was not possible to take the hydrolytic retention times of the Matero wastewater stabilization ponds into account. If the retention times for the wastewater could have been estimated, the removal efficiency of the studied pharmaceuticals in the wastewater stabilization ponds could have been calculated, and this would have added depth to the study. However, even without knowing the removal efficiency it seems evident that the ponds do not function to their full capacity and removal of pharmaceuticals from the wastewater is minimal.

Ethical questions were discussed with the University of Zambia personnel who required applying for an Ethical Clearance for the work as the study plan included taking samples of human urine, even if anonymous. In the process it was decided that urine should be collected only from public toilets. In the end, the description of a public toilet needed to be stretched as there simply were not enough actual public toilets in use to take samples from: ‘public’ was considered to mean toilets used by more than one household and more than 10 people. This did not endanger the anonymity of the study results.

During the data collection it became evident that obtaining reliable data on the consumption of medicines was not always possible, especially in the case of often sensitive HIV medications. For instance only one household from whose shared UDDT a sample was taken from, reported the use of ART, but based on the results it can be deducted that this was not the only household where antiretrovirals were being used on a daily basis. It was important to pay attention on communicating about the aim and content of the study, interviewing the local people for the background surveys and presenting the research results, as the study subject was of sensitive nature. It needed to be ensured that households with people living with HIV could not be identified at any point, especially as the study area was rather small, close and densely populated. Therefore the sample types are not specified for instance in the sample map of Picture 7. in order to inhibit identification of households with high usage levels of pharmaceuticals, especially antiretrovirals.

6 CONCLUSIONS

Fertilizer use of the source-separated urine would be a responsible form of agricultural production reducing the need for finite mineral fertilizers, but in locations with high disease prevalence and corresponding consumption of medicines, protective measures would be essential in order to enable safe agricultural use of urine. The high concentration of pharmaceuticals detected in source-separated urine in this study calls for careful considerations to be made when the urine is intended to be directly used as a fertilizer for food crop production. Unfortunately, the promising nutrient cycling potential of human urine is severely compromised if pharmaceutical residues are present in the source-separated urine, however the high nutrient values can still be taken into use with effective treatment of the source-separated urine. Appropriate treatment options for removal of pharmaceutical traces could be for instance electrodialysis, struvite precipitation, nitrification and solar evaporation, electrolysis and a combination of nitrification and distillation (Pronk & Koné 2009; Udert et al. 2015).

The sanitation infrastructure of the study area is insufficient, but also the collection and storage infrastructure for the source-separated urine produced in the study area are not in balance with the production volumes or with the currently recommended storage period of ≥6 months in order to ensure microbiological safety of the urine used as a fertilizer (WHO 2006). The demand for fertilizer in the study area is also currently lower than the production capacity of urine, which leads to unsafe disposal of the source-separated urine and spreading of the nutrients and pharmaceutical contamination in the environment. Studies have shown that the recommended ≥6 month storage period is also not effective in degrading the pharmaceuticals excreted in the urine and even urine stored appropriately is not necessarily safe for agricultural application (Jaatinen et al. 2016). The measured high pharmaceutical concentrations in the source-separated urine can therefore pose an impediment to the agricultural use of urine collected from Madimba and most probably also from other similar settings with high disease prevalence and corresponding consumption of medicines. As even low doses of pharmaceutical contamination in the environment can lead to development of antimicrobial resistance through easily disseminated antimicrobial resistance genes ARGs, precautionary measures are recommended even if the knowledge is still limited.

Since only urine from households with no medication use could be appointed for direct fertilizer use without further treatment, implementing such household-dividing measure would exclude and pose a risk of stigmatization to households living with HIV and other diseases requiring daily medication, and would therefore prove problematic. It could also lead to dishonesty about the use of medication in order to avoid stigma, which could compromise the qualities of the urine intended for fertilizer use because of the potential pharmaceutical contamination. Therefore such measure cannot be recommended in practice.

The environment, especially the vulnerable aquifers of Lusaka, and human health should nevertheless be protected with usage of the already existing urine-diverting dry toilets (UDDTs) and the source-separation of urine in the study area. Source-separation of urine would confine the excreted pharmaceutical contaminants into manageable volumes that could be either treated for pharmaceutical reduction before being used or sold as a fertilizer or disposed of appropriately. Currently the UDDTs built in the study area are often not used appropriately, as due to occurred challenges many of them are either not used at all or they are used in the same manner as pit latrines. In case of the small number of the UDDTs where urine separation is still taking place, the urine container is often unsafely emptied on the ground, which further spreads pharmaceutical contamination in the environment. The importance of the source-separation of urine together with successful local ownership of the UDDTs should be carefully understood and ensured in this and similar development projects. Also collection, transport and safe usage or disposal of the source-separated urine should be effectively and appropriately organized in order to protect the environment and human health. If this urine could be treated for effective pharmaceutical residue removal and reused as a fertilizer for either food or non-food crops, it would have potential to enhance local agriculture, nutrient recycling and business opportunities.

The findings of this study suggest that whether fertilizing food crops with source-separated urine is a viable and safe option should be evaluated in relation to the local context. Taking into account the local consumption of medicines and the local water and sanitation systems through which the urine and the related pharmaceutical residues flow is of utmost importance. Especially in the informal settlements and peri-urban areas of the developing world where on-site water and sanitation systems are common, it should be noted that the hydrological cycles and the agricultural systems are easily exposed to these trace level pollutants. Moreover, it is often the most vulnerable populations that are affected by the potential risks associated with pharmaceutical contaminants in the environment such as antimicrobial resistance.

The field of international development cooperation has over the years seen enough ‘one-size-fits-all’ solutions, where a measure that works in the global North is implemented in the global South, without taking the local context sufficiently into account. In case of promoting the use of source-separated urine as a fertilizer for food crops, the local residents of Madimba hesitating with the full adoption of the practice may have been on the button, for one reason or another, and this is one of the important lessons learned from this study. The high prevalence of HIV and other diseases have a direct effect on the viability of agricultural use of source-separated urine in an area characterized by such factors, and while designing development projects around this subject, it is essential to apply the best available knowledge taking even the smallest particles in the whole picture into account, in this case the pharmaceutical traces acting as micropollutants. Also following the precautionary principles of environmental and social protection is necessary for management of risks associated with these contaminants.

Cycle thinking is the key to sustainable development in a world of delicate and intertwined life cycles, but in order to promote it, we must also ensure the long-term safety of our chains of action.

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APPENDICES

UNIVERSITY OF JYVÄSKYLÄ

WATER SAMPLE INFORMATION FORM

SAMPLING INFORMATION **SAMPLE NO.**

Name of sampler:	Volume of sample:
Bottle type:	Place:
Plot no:	GPS: S E
Date:	Time:
Well type: ☐ Shallow well: P / N ☐ Borehole ☐ Stream ☐ Other:	
Sample taken: ☐ Directly from well ☐ Through a pump ☐ Tap ☐ Bucket ☐ Other:	
Filter used? Yes / No Type:	Age of the well:
Well last emptied and cleaned:	Well last disinfected:
No. of people using the well:	Animals drinking from the well? Yes / No
Changes in water quality due rain? Yes / No	Surface water entering the well? Yes / No
Distance to ditch / stream: m	Depth of the well: cm Diameter: cm
Estimated volume of water:	Water used for drinking? Yes / No
Distance to: ☐ Dry toilet m ☐ Pit latrine m ☐ Urine-fertilized garden m Elevation and soil type:	
Distance to: ☐ Animal shelter m ☐ Manure-fertilized field m ☐ Other: m Elevation and soil type:	
Other environmental factors (industry, road, dump etc.):	
Notifications on water (color, smell, turbidity etc.):	
Date:	**Signature:** Printed name:

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URINE SAMPLE INFORMATION FORM

SAMPLING INFORMATION **SAMPLE NO.** ☐

Name of sampler:	Volume of sample:
Bottle type:	Place:
Address:	Coordinates:
Date:	Time:
Toilet type: ☐ UDDT ☐ Other:	
Sample taken: ☐ From a container ☐ Other:	
Urine stored: ☐ In a container ☐ Other:	
Storing date:	Length of storage:
Storing facility description:	
Toilet is: ☐ Private ☐ Public ☐ School ☐ Other:	
No. of people using the toilet:	Shared between families? Yes / No
Used medication:	
Distance from toilet to: ☐ Shallow well m ☐ Borehole well m ☐ Ditch / stream m Elevation and soil type:	
Toilet description:	
Notifications on urine (color, turbidity etc.):	
Date:	**Signature:** Printed name: