Towards an Improved Ceramic Water Filter Paradigm: From Design Optimization to Participatory Implementation

by

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Abstract

Rural and low-income populations in sub-Saharan Africa (SSA) are disproportionately impacted by challenges with safe drinking water access. Significant attention has resultingly been directed towards home-based drinking water treatment as an immediate-term solution. Ceramic water filters (CWFs) are one such technology that is widely promoted due to its simplicity in terms of use, maintenance, and manufacture. However, cost, flowrate, breakage, and general misuse/disuse in the field are significant barriers to proliferation, adoption, and associated impact. This research addresses each of these limitations by evaluating technological innovations and participatory implementation in rural Tanzania.

CWFs are typically produced by mixing clay, sawdust, water, and silver nanoparticles (AgNPs), pressing the mixture into a pot shape, and firing it in a kiln. AgNPs are added to enhance disinfection but are among the most expensive of filter elements. Cost reduction is thus explored through investigation of AgNP replacement and/or supplementation with zinc oxide (ZnO), an inexpensive alternative. Metals are challenged in isolation and combination against *E. coli* within batch and filter phases and disinfection and elution are assessed across time and varying water qualities. Combined AgNP-ZnO treatment proved synergistic under all conditions, consistently outperforming either metal alone. Maximal effects are further observed when zinc concentrations above 200 ppb are combined with silver concentrations above 10 ppb within the filtrate. Supplementing silver with zinc in CWFs can therefore simultaneously reduce cost while improving bactericidal efficacy.

Input sawdust size and proportion within a CWF mixture is also related to flowrate, flexural strength, and bacteria removal, yet the nature of these relationships is unclear. Modelling these performance measures is consequently challenging, especially in such a way that is

scalable between manufacturing settings. CWF disks with varied input material characteristics are therefore evaluated for flow, strength, and disinfection efficacy. Nested stepwise multiple regression analysis with possibility-based design optimization (PBDO) is then investigated as a novel approach to predicting filter performance with generalized porous media characteristics. Optimum material parameters that maximize CWF flowrate while maintaining sufficient bacteria removal and strength are also identified. Results show that these key performance metrics are modeled well by porosity, density, and intrinsic permeability, successfully demonstrating the validity of this method. The PBDO output further estimates that flowrate may be increased to above 8 L/hr without compromising water treatment capacity or strength if is porosity is <48%, dry density is >1.2 g/cm^3, and intrinsic permeability is <29 x 10^{-9} cm^2.

Finally, multi-year community engagement and participatory program development processes in Longido, Tanzania are described. Three knowledge communication and CWF provision structures implemented in collaboration with a partner non-profit and other local stakeholders are subsequently evaluated. Filter adoption, breakage, and associated health are monitored over 27 months. Multi-week and repeated knowledge communication proved critical to promoting CWF usage, maintenance, and protection. The study therefore shows that long-term participant engagement and support is necessary for CWFs to have desired positive outcomes; one-off filter and knowledge provision is insufficient to meet user needs.

In summary, this dissertation demonstrates harmonious innovation and synthesis of technical and social factors related to CWF sustainability from the laboratory to the field, advancing progress towards improved filter performance, usage, and longevity.

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List of Acronyms and Symbols

Ag	Silver
Ag_2O	Silver Oxide
Ag_2S	Silver Sulfide
AgCl	Silver Chloride
AgF	Silver Fluoride
$AgNO_3$	Silver Nitrate
AgNP	Silver Nanoparticle
AgO	Silver Oxide
AHP	Analytical Hierarchy Process
Al_2O	Aluminum Oxide
AT	Appropriate Technology
BPEI	Branched Polyethyleniminie
BSD	Big Sawdust Particle Size
C:SD	Clay-Sawdust Ratio
CAWST	Centre for Affordable Water and Sanitation Technology
CCD	Central Composite Design
CCME	Canadian Council of Ministers of the Environment
CFD	Computational Fluid Dynamics
CFU	Colony Forming Units
CI	Confidence Interval
Cl^-	Chloride
CMWG	Ceramics Manufacturing Working Group
C_o	Initial Concentration

COSTECH	Tanzanian Commission on Science and Technology
COV	Coefficient of Variance
C_t	Concentration at Time, t
Cu	Copper
$Cu(NO_3)_2$	Copper Nitrate
CuO	Cupric Oxide
CUREB	Carleton University Research Ethics Board
CWF	Ceramic Water Filter
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
DSLR	Digital Silngle-Lens Reflex
E	Young's modulus
e^-	Free Electron
E. coli	Escherichia Coli
EDS	Energy Dispersive Spectrocsopy
EtOH	Ethanol
FC-CCD	Face-Centered Central Composite Design
FM	Firing Material
FR	Flowrate
g	Graviatational Force
GNP	Gross National Product
h^+	Positive Hole
HCG	Health Clinic Group
HCl	Hydrochloric Acid
hrs	Hours

Hs	Enthalpy
HSD	Honest Significant Difference
IBRD	International Bank of Reconstruction and Development
ICP-MS	Inductively Coupled Mass Spectrometry
IPCC	International Panel on Climate Change
IS	Ionic Strength
IT	Intermediate Technology
IWA	International Water Association
JMP	Joint Monitoring Program
K	Hydraulic Conductivity
KH_2PO_4	Potassium Phosphate
KLG	Kimokouwa Literacy Group
LDHC	Longido District Health Center
LRV	Log Removal Value
MDG	Millennium Development Goal
MDP	Maasai Development Program
ME	Metal Eluted
$MgCl_2$	Magnesium Chloride
MgO	Magnesium Oxide
mM	Milimolarity
MNP	Metal Nanoparticle
MOR	Modulus of Rupture
MS	Microsoft
Na_2HPO_4	Disodium Phosphate
NaH_2PO_4	Soldium Phosphate

$NaNO_3$	Sodium Nitrate
NaOH	Sodium Hydroxide
NGO	Non-Governmental Organization
NM-AIST	Nelson Mandela African Institute of Science and Technology
N_o	Initial Concentration
NOM	Natural Organic Matter
NSERC	Natural Sciences and Engineering Council of Canada
N_t	Concentration at Time, t
OH^-	hydroxide
OLG	Oldorko Literacy Group
P	Load
P. Putida	Pseudomonas Putida
PBDO	Possibility-Based Design Optimization
PDF	Probability Distribution Function
PEPAM	Programme d'Eau Potable et d'Assainissement du Millénaire au Sénégal (Millennium Water and Sanitation Program)
PfP	Potters for Peace
PHAST	Participatory Hygiene and Sanitation Transformation
pKsp	Solubility Constant
PMA	Performance Measure Approach
POU	Point of Use
POUWTS	Point of Use Water Treatment Solution
ppb	Parts per Billion
PVP	Polyvinylpyrollidine
Q	Flowrate

QA	Quality Assurance
QC	Quality Control
r	Radius
RBDO	Reliability-Based Design Optimization
RMSE	Root Mean Square Error
ROS	Reactive Oxygen Species
RTI	Research Triangle Institute
SA	Surface Area
SAP	Structural Adjustment Program
SARAR	Self-Esteem, Associative Strengths, Resourcefulness, Action-Planning, and Responsibility
SD	Standard Deviation
SDG	Sustainable Development Goal
SE	Standard Error
SEM	Scanning Electron Microscopy
-SH	Thiol
SLSQP	Sequential Least Squares Programming
SMDWS	Safely Managed Drinking Water Services
SoDIS	Solar Disinfection
S_s	Entropy
SSA	Sub-Saharan Africa
SSD	Small Sawdust Particle Size
T	Temperature
TEMBO	Tanzanian Education and Micro Business Opportunity
TiO_2	Titanium Dioxide

TZS	Tanzanian Shillings
UN	United Nations
UNCED	UN Conference on Environment and Development
UNGA	United Nations General Assembly
UNICEF	United Nations Children's Fund
USAID	United States Agency for International Development
USEPA	United States Environmental Protection Agency
USSR	United Socialist Soviet Republic (Soviet Russia)
v	Velocity
W2W	Wine to Water International
W2WEA	Wine to Water East Africa
WaSH	Water, Sanitation, and Hygiene
WBC	WaSH-related Behaviour Change
WHO	World Health Organization
WWI	World War I
WWII	World War II
XRF	X-Ray Fluorescence
Zn	Zinc
ZnO	Zinc Oxide
z_o	Height of Water Above Filter Base
μ_w	Viscosity of Water
Ø	Diameter
κ	Intrinsic Permeability
λ	Thermal Conductivity
ρ	Density

Υ	Interfacial Surface Energy
$\boldsymbol{\varphi}$	Porosity

Thousands have lived without love,

not one without water.

- *W.H. Auden*

Chapter 1: Introduction

1.1 Water in a Changing Climate

Water is a primary vehicle of planetary function. It has sustained life, maintained ecosystems, and facilitated all material creation for time immemorial. The ongoing and accelerating impacts of climate change are, however, jeopardizing this delicate balance. Rising temperatures are decreasing both the quantity and quality of freshwater reserves, exacerbating the already vast inequalities in water access for the continuation of human and non-human life alike (Ceballos and Ehrlich 2018). Increasingly erratic weather patterns are producing more extreme and less predictable rainfall episodes that threaten to overwhelm existing infrastructure, limit capacity to manage water volumes over time, spread greater contamination throughout waterways, and cause embodied damages (Bates, et al. 2008). Meanwhile, population growth is outpacing freshwater replenishment (World Bank 2021). Demand is growing while supply is reducing in both quality and quantity. With concurrent social, political, and economic inequalities, as well as worsening health, financial, and ecological crises, current trends point to a future of exacerbated marginalities and inequal exposures to embodied risks (Hallegatte, et al. 2016, GBD Risk Factor Collaborators 2020). Identifying means of mitigating these risks is moreover among the most pressing challenges of the 21st century.

One geography which further illustrates systematic inequity and water-related vulnerability is rural sub-Saharan Africa (SSA). Of the nearly two billion individuals globally who lack access to safely managed drinking water services, more than half reside only within these

areas (World Bank 2021). Similarly, of the more than 500,000 children that die from diarrheal illness each year, caused, in large part, by unsafe water consumption (WHO 2017), more than 60% are from SSA (GBD Risk Factor Collaborators 2020), of whom 75% are estimated to reside in rural areas (Yaya, et al. 2019). Meanwhile, the International Panel on Climate Change (IPCC) predicts that these same locations will experience among the most severe climate impacts within this century, threatening water safety and security, as well diverse other environmental factors contributing to social and economic welfare (Christensen, et al. 2007). Progress towards improved safe water access within rural SSA is urgently required.

1.2 Problem Statement

Particularly since the Millennium Development Goals (MDGs) in 2000, significant discursive and research attention has been directed towards point-of-use water treatment solutions (POUWTS) (Sobsey, et al. 2008, Murphy, McBean and Farahbakhsh 2009). These technologies are varied in size, cost, and water treatment capability, though are generally decentralized, small in scale, and able treat water at the point of consumption. They are thus immediately implementable and significantly more affordable than their conventional centralized counterparts (Santos, Pagsuyoin and Latayan 2016). Diverse scholars and international institutions have promoted their inclusion within international water, sanitation, and hygiene (WaSH) programming, especially for rural and remote contexts (WHO; UNICEF 2017). That is, POUWTS are often recommended when superior centralized water treatment infrastructure is infeasible in the immediate term. The means of selecting a POUWTS for a given program is further an area of active research commonly referenced as appropriate technology (AT) identification (see Chapter 3) (C. P. Sianipar, et al. 2013).

Briefly, considerations involve matters such as resource accessibility, ease of manufacture, ease of use, ease of maintenance, and energy consumption, among others (see Chapter 3).

Within this body of literature, ceramic water filters (CWFs) are recognized as of the most effective and preferable to users (Clasen, et al. 2015, Burt, et al. 2017, Santos, Pagsuyoin and Latayan 2016). They are manufactured by combining clay, a firing material (FM; e.g., sawdust), silver nanoparticles (AgNPs), and water into a homogeneous mixture, which is then dried and subsequently fired in a kiln to burn away the FM and produce a porous matrix (CMWG 2010). CWFs are therefore primarily composed of materials easily sourced across diverse geographies and may be manufactured at the location of requirement. They also widely meet or exceed international drinking water guidelines for household-scale filtration technologies (WHO 2011), have a relatively simple use and maintenance protocol, and cost less than many POUWTS alternatives (Sobsey, et al. 2008, Pagsuyoin, et al. 2015). CWFs have thus been distributed within diverse geographies throughout the global South, and particularly within rural areas (van Halem, van der Laan and Soppe, et al. 2017, Clasen, et al. 2015). They are further recommended by scholars and WaSH-focused institutions as a critical technology for addressing a lack of access to safe drinking water in the immediate term (UNICEF 2016, CAWST 2020, Rayner, et al. 2017).

Some significant challenges, however, continue to hinder acceptability and proliferation. Most notably, despite being generally more affordable than other POUWTS, CWF cost is often reported to exceed the means of intended users (Luoto, et al. 2012, Burt, et al. 2017, Santos, Pagsuyoin and Latayan 2016). The inclusion of AgNPs within CWF fabrication, though widely accepted as valuable for providing additional disinfection efficacy, can further significantly increase the cost of manufacture (Oyanedel-Craver and Smith 2008, van der

Laan, et al. 2014, CMWG 2010). Research is therefore needed to identify means of reducing the quantity of AgNPs such that CWFs are more affordable to target users living in low-income and resource-restricted communities while still treating water sufficiently. Investigation of Ag replacement and/or supplementation with an alternative metal nanoparticle (MNP) species with a lower cost may offer a novel means of improving accessibility. However, few studies have evaluated CWF impregnation with non-Ag metals. Further, of those available, most are focused on cupric oxide (CuO) or iron oxide, which offer limited differences to silver in terms of cost reductions (Brown and Sobsey 2009, Jackson, Smith and Edokpayi 2019, Lucier, Dickson-Anderson and Schuster-Wallace 2017). Meanwhile, zinc oxide (ZnO) has well-documented bactericidal capacity, efficacy in treating and/or preventing diarrheal illness, and is commonly available at a very low cost (CCME 2018, Malik, et al. 2013, Sirelkhatim, et al. 2015). Yet ZnO disinfection in water treatment applications remains highly understudied, with particularly limited investigation of its inclusion within CWFs (Huang, et al. 2018, Dimapilis, et al. 2018).

An additional challenge to CWF sustainability is flowrate, as filtrate production efficiency is commonly reported as insufficient to meet household drinking water demands (Annan, et al. 2014, Yakub, et al. 2013, Rayner, et al. 2017). FM proportion and size are furthermore regarded as the key determinants of flow, as FM burns away to form pores that allow water to pass through the matrix (CMWG 2010). Meanwhile, FM proportion and size are also related to disinfection capacity, as pore size exclusion is the primary mechanism of microbe removal. Finally, these same input parameters impact material strength, and by extension, CWF longevity, as filter breakage after deployment is a leading cause of disuse (Soppe, et al. 2015, Lemons, et al. 2016, Brown, Proum and Sobsey 2009). An optimal combination of

these input material parameters may therefore exist which maximizes flowrate and material strength while maintaining microbe removal performance above drinking water guidelines. And while some research has evaluated the impacts of varying these material parameters on performance (Soppe, et al. 2015, Servi, et al. 2013, Rayner, et al. 2017), no studies are known to have identified optimum metrics that may scale across manufacturing facilities. In other words, correlation between these key performance metrics and input materials is often unique to the facilities within which CWFs are manufactured. Research investigating means for comparing CWFs across factories, as well as identifying optimum inputs, is thus needed.

A perfectly cost-effective and materially robust technology will still not necessarily improve safe water access after deployment though (C. P. Sianipar, et al. 2014). Diverse scholarship and practical experiences have highlighted that CWF usage and maintenance decreases over time once within a household, potentially limiting impacts on associated health outcomes (Martin, et al. 2018, Marshall and Kaminsky 2016). Further, with filter breakage remaining a particularly acute challenge, technological longevity may also involve a behavioral component. CWF value is therefore not solely a matter of its ability to meet technical specifications but also its ability to be integrated into the lives of its users. Summarily, the provision of safe water access does not manifest from the provision of technology but the provision of programming that facilitates usage of that technology. How to do so is, however, unclear and is further a matter of active debate (Fiebelkorn, et al. 2012, Kraemer and Mosler 2010, Bishoge 2021). For instance, the United Nations (UN) Sustainable Development Goal (SDGs) 6.a directs implementers to incorporate "community participation" to facilitate improved uptake of WaSH programs (UN 2021). But *what is* participation in practice, and how does it translate to water-related health outcomes? Similarly, scholars have highlighted

a need to move away from technological provision models to ones which incorporate WaSH knowledge communication (Marshall and Kaminsky 2016, Dreibelbis, et al. 2013). However, valuable methods related to the type and duration of such communications are not well elucidated. As such, while principles for POUWTS implementation, and CWF implementation specifically, are present within WaSH discourse, a significant gap exists regarding how these principles are applied. Research is therefore required to identify CWF intervention and knowledge communication methods that promote community participation, technological usage, and associated positive health outcomes within a rural SSA context.

This research moreover combines technical explorations of methods for innovating CWFs with investigation of technological implementation in rural Tanzania. Specifically, ZnO is examined as an AgNP supplement under batch conditions and when applied to a CWF to determine its appropriateness as a cost-reducing agent. Nested multiple regression analysis and machine learning is also used to study, model, and optimize CWF input material parameters and porous media characteristics such that outcomes may be scaled across manufacturing facilities. And finally, participatory and community-driven methods for CWF distribution and knowledge communication are evaluated with respect to their impacts on technological usage, social determinants of environmental health, and associated water-related health outcomes in Longido, Tanzania.

1.3 Location Profile: Longido, Tanzania

Longido is the northernmost district of the Arusha Region of Tanzania in East Africa. It is home to a total population of approximately 120,000 inhabitants, disparately residing at a density of approximately 15/km^2 (Brinkhoff 2012, Worldometer 2020). The climate is semi-arid with a diurnal rain season that typically occurs between February-April and October-

November. Approximately 350 mm of collectable rain falls each year, the vast majority of which is during the first wet season. Residents primarily collect their water from village taps fed by basins collecting runoff atop Mount Longido, or since 2020, pipelines transporting water from the tropical Kilimanjaro region approximately 250 km south. Alternatively, some individuals collect rainwater directly or may access dugout wells, shallow wells (where available), or water collected in an earthen dam colloquially known as "the Ocean". No water sources in this area are treated and diarrheal illnesses are widely considered a significant challenge (Mshida, et al. 2017). Water quality and quantity are thus both significant challenges for residents of this geography.

Within this research, CWF implementation is evaluated primarily within the Kimkokouwa and Orbamba wards, highlighted in green in Figure 1.1. Participants in this work were further women who identify as Maasai and follow pastoralist livelihoods. Common activities among participants and their communities include livestock keeping, beadworking, and living in acacia-encircled compounds called bomas in which homes are walled by clay and cow dung and roofs are made of brush.

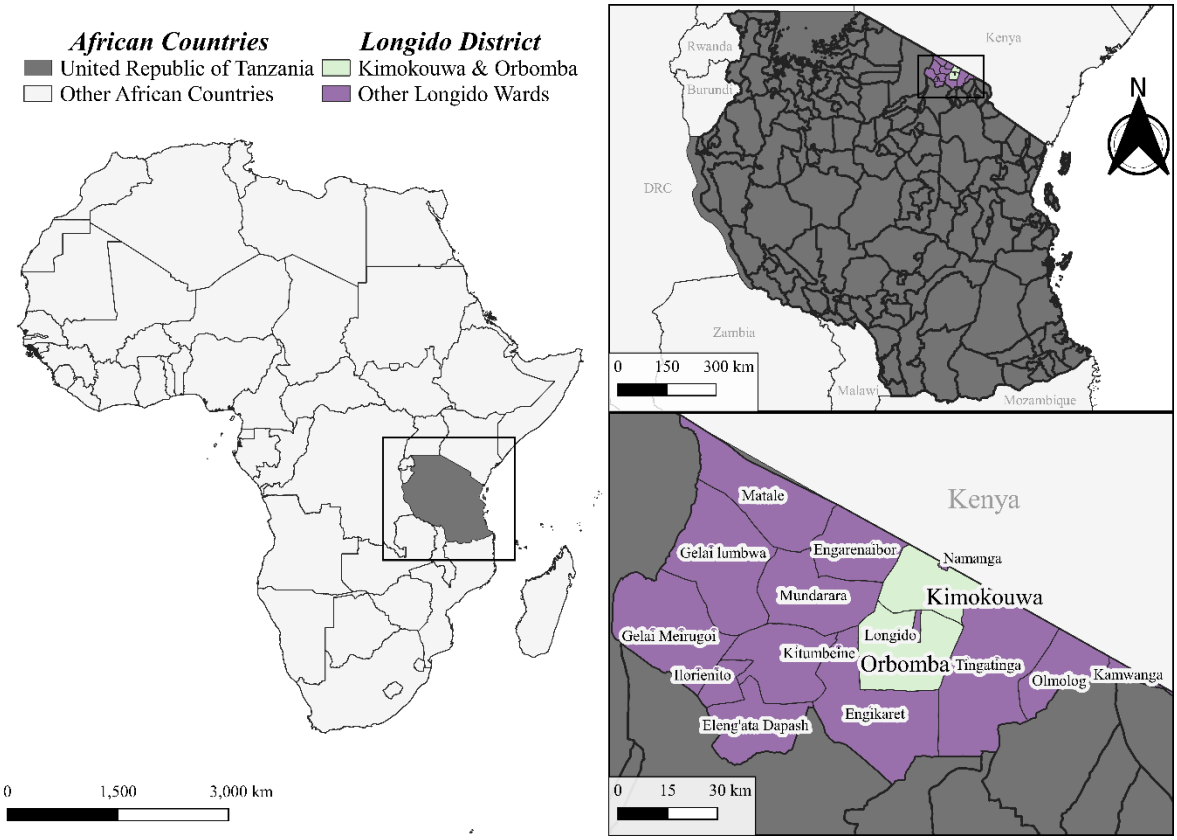

Figure 1.1 Longido District of Northern Tanzania in East Africa (Data: (HumData Exchange 2018))

1.3.1 A Brief History of Maasai Communities in Tanzania

Maasai in Tanzania are a self-identifying Indigenous population who follow a 'pastoralist livelihood' and have resultingly been subject to systemic discrimination and marginalization since the British empire (D. Hodgson 2011a, 2011b, 2011c). For instance, one British administrator described Maasaidom as "a beastly, bloody system, founded on raiding and immorality" (Rigby 1992a). Later, a once-described "Maasai expert" wrote, "the traditional Maasai social structure and cultural institutions fundamentally constrain development initiatives" (Evangelou 1984). And more recently, a prolific Maasai political advocate referred to government programs as "trying to 'phase out' pastoralism as a livelihood" (D. L. Hodgson 2011c). Tanzanian Maasai have continually fought for the survival of their

traditions and modes of production, often resulting in conflicts with colonial modernization efforts and postcolonial neoliberal agendas.

Specifically, before British rule in Tanzania and Kenya, the Maasai were but diverse peoples with a shared lingual history (Maasai meaning 'speakers of the Maa language') and practice of a semi-nomadic pastoralist tradition (D. Hodgson 2001a). Then, in 1922, "the British strove to consolidate and bound people into distinct categories – 'tribes' – then place these tribes within demarcated, controllable spaces. Once ethnic identity became spatially enclosed and bounded, colonial administrators could establish and expand their territorially based system of political control through recognition and collaboration with the 'chiefs' of these 'tribes'" (D. Hodgson 2001a). The newly created Maasai 'tribe', earlier categorized thusly according to, in part, their fluidity in movement to find food, water, and grasslands for cattle to graze, was thus resultingly constricted and ethnicized accordingly.

Maasai communities across Tanganyika subsequently lost access to their ancestral lands, and by extension, access to these critical physical resources (particularly in dry seasons); indeed, the desire to regain the right to mobility remains an active element of Maasai political advocacy (D. L. Hodgson 2011c). That is, colonists actively worked to promote sedentism among Maasai populations such that these communities would adopt Western-style animal husbandry practices and associatively *modernize* to participate in global capitalism (Rigby 1992b, D. Hodgson 2001a). These notions then extended throughout decolonization and into the 21st century, as national and international development programs coerced populations into behaviours and modes of production that more closely resembled those in higher-income

countries[1]. Simultaneously, increasing mining, agriculture, and tourist operations continually reduced available grazing lands, forcing Maasai populations onto smaller and smaller parcels of land for grazing and resource access[2] (D. Hodgson 2001b, D. L. Hodgson 2011c, Azarya 2004, Gardner 2012). Today, these exercises have resulted in continued Maasai marginality and disproportionately unequal health and economic outcomes (Lawson, et al. 2014).

1.4 Research Objectives and Scope

This research has three overall research objectives, which each include specific objectives towards their accomplishment:

1. Reduce CWF cost through evaluation and minimization of AgNP impregnation
    a. Investigate AgNP impregnation on impacts on CWF bacteria removal
    b. Investigate AgNP disinfection efficacy across concentrations and water qualities
    c. Investigate ZnO as an alternative and/or supplement to AgNPs for impregnation within CWFs
    d. Investigate the microbial behaviour of CWF effluent during long-term storage when impregnated with varying concentrations of AgNPs or ZnO
2. Characterize CWF material parameters and related performance to identify an optimum design that maximizes flowrate, flexural strength, and bacteria removal

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[1] A particularly outrageous example is "Operation Dress-Up", a national initiative within the 1970s that aimed to incentivize men to wear shirts and trousers, women to stop wearing traditional beaded necklaces, and all to refrain from using red ochre for hair and body painting. It was specifically promoted as a health project to "assist the Maasai to attain a level of development equal to that of the rest of the people in the country" (Schneider 2006). The program, however, had no grounding in science, especially considering ochre is used mostly to prevent lice.

[2] Events which continue today, as seen in Loliondo, a district adjacent to Longido (UNHR-OCHR 2022)

a. Measure impact of variable Clay-FM proportions and FM sizes on flowrate, flexural strength, and bacteria removal.
b. Develop generalized models to predict performance metrics from measured characteristics of porous media
c. Use machine learning and uncertainty analysis to identify range of possible input, material, and performance optimums

3. Investigate participatory WaSH education and CWF implementation model to determine key factors for sustained technology usage and associated positive health outcomes in Longido District, Tanzania
   a. Develop locally oriented and user centered CWF distribution and WaSH knowledge communication program within study community
   b. Evaluate impact of CWFs and knowledge communication on associated water-related health outcomes and social determinants of environmental health
   c. Evaluate the impact of knowledge communication duration and frequency on associated water-related health outcomes and social determinants of environmental health.

1.5 Thesis Structure

This thesis is divided into four sections: Introductions, Technical Research, Field Research, and Reflections. The chapters were written in journal article format, with Chapters 2, 4, 5, 6, 7, and 8, each representing one research or literature review article. Chapters 3, 9 and 10 are, however, not presented as publications but individual pieces to complement the other chapters herein. Chapter topics are as follows:

Chapter 1, Introduction: introduces the research's problem statement and the objectives identified to address it.

Chapter 2, Literature Review – Technical (Published): provides background information relevant to the technical engineering components of this research.

Chapter 3, Literature Review – Field & Social Sciences: provides background information relevant to the social scientific components of this research.

Chapter 4, Technical Phase I Results (Published): presents the results of the first stage of this research, which investigates the kinetics and synergistic bactericidal efficacy of AgNPs and ZnO when combined under batch conditions at 1 mg/L total concentrations. The study also includes a preliminary investigation of combined silver and zinc impregnation into CWF pots, where bacteria removal is measured immediately after filtration, as well as after 24 hours of storage.

Chapter 5, Technical Phase II Results: presents the results from the second stage of this research, which explores the disinfection efficacy of AgNPs and ZnO when released from a CWF disk (scaled-down CWF). Metal release values are enumerated and correlated with changes to bacterial concentrations immediately after filtration, as well as over 72 hours of storage. Eluted concentrations were also replicated under batch conditions to investigate the specific impacts of Ag and Zn during storage. Chick-Watson kinetic analysis is used for comparison.

Chapter 6, Technical Phase III Results: presents the third and final stage of the technical engineering research, which investigates the impacts of varying clay-sawdust (C:SD) ratios and sawdust particle sizes on CWF disk flowrate, bacteria removal, and material strength. A

face-centered central composite design (FC-CCD) experimental structure is used, and nested multiple regression models are developed to predict the abovementioned performance metrics. Possibility-based design optimization (PBDO) is used to identify optimum filter parameters under deterministic, safest-case, and best-case scenarios to account for uncertainty.

Chapter 7, Social Sciences Phase I Results (Published): presents the first stage of the field research conducted within this study, where the impacts of distributing CWF pots to 50 households across the Kimokouwa ward of the Longido district in rural Tanzania were evaluated. Weekly participatory WaSH education programming was repeated cyclically over an 18-month evaluation period and child and maternal diarrheal health, filter breakage, and knowledge retention were evaluated. The second phase of this research is not prepared within a publication format, but preliminary results may be found within Appendix B.

Chapter 8, WaSH Policy/Program Review & Analysis, Reflections (Submitted): is the final stage of the field research program, providing an historical analysis of policy, programming, and discourse within the WaSH sector since 1918. The study specifically identifies key moments and trends that have produced the current paradigm of WaSH programming and research centered around facilitating behaviour change among recipients. Evaluation of history's influence on resulting intersubjectivities within international WaSH is then presented, followed by some recommendations on reform. This article may be considered my reflexive response to some of the lessons learned conducting WaSH research in a rural Tanzanian context. The paper was submitted and returned with major revisions, which have been completed; a final revised version is due after this defense.

Chapter 9, Summary and Experiential Learnings, Reflections: is a short exploration of my thoughts and feelings about conducting this research and my ability to advance progress *towards and improved ceramic water filter paradigm*. Departing from traditional academic knowledge communication, this section is intended as a reflexive and informal piece to accompany Chapter 8.

Chapter 10, Conclusions & Recommendations: summarizes this research and provides concluding recommendations for future work.

References: lists all references sourced within this thesis.

1.6 Articles Summary and Authors Contributions

Author contributions presented below follow the Elsevier CRediT convention.

(Chapter 2) Article 1: Mechanisms and Efficacy of Disinfection in Ceramic Filters: A Critical Review

Authors: Robbie A. Venis[1], Onita D. Basu[1]

Published: Critical Reviews in Environmental Science and Technology

[1] Department of Civil and Environmental Engineering, Carleton University, 1125 Colonel by Drive, Ottawa, ON, K1S 5B6, Canada

Author Contributions:

Table 1.1 Article 1 Author Contributions

Contribution	Author 1: Robbie A. Venis	Author 2: Onita D. Basu
Conceptualization	X	X
Methodology	X	
Software		
Validation		X
Data Analysis	X	X
Investigation	X	
Resources		X
Data Curation	X	

Writing – Original Draft	X	
Writing – Review & Editing	X	X
Visualization	X	
Supervision		X
Project Administration		
Funding Acquisition		X

(Chapter 4) Article 2: Silver and Zinc Oxide Nanoparticle Disinfection in Water Treatment Applications: Synergy and Water Quality Influences

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Software		
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Formal Analysis	X	
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Resources		X
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(Chapter 5) Article 3: Elution and Disinfection of Silver and Zinc Nanoparticles in Co-Fired Ceramic Water Filters

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Investigation	X	
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Data Curation	X	
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Visualization	X	
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(Chapter 6) Article 4: Ceramic Water Filter Material Design Optimization

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Software	X	X	
Validation		X	X

Formal Analysis	X	X	
Investigation	X		
Resources			X
Data Curation	X		
Writing – Original Draft	X		
Writing – Review & Editing	X	X	X
Visualization	X		
Supervision			X
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(Chapter 7) Article 5: Towards a Participatory Framework for Improving Water & Health Outcomes: A Case Study with Maasai Women in Rural Tanzania

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Methodology	X	X	X	X	X	
Software						
Validation		X				X
Formal Analysis	X					
Investigation	X		X	X	X	
Resources	X					X
Data Curation	X					
Writing – Original Draft	X					
Writing – Review & Editing	X	X			X	X
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(Chapter 8) Article 6: Behaviour Change in Water, Sanitation, and Hygiene: A 100-Years Perspective

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Data Curation	X
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Writing – Review & Editing	X
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Supervision	
Project Administration	X
Funding Acquisition	

Chapter 2: Technical Literature Review

Mechanisms and Efficacy of Disinfection in Ceramic Water Filters: A Critical Review

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Abstract:

Diarrheal illnesses claim the lives of hundreds of thousands of children each year, most of whom live in rural and low-income communities. Increasing access to safe drinking water would significantly impact this pervasive challenge, and Ceramic Water Filters are regarded as one water treatment technology with the potential to do so. Silver is commonly added to CWFs to improve disinfection; however, it also significantly adds to the technology's cost. Therefore, a thorough understanding of silver disinfection efficacy and disinfection mechanisms in relation to CWFs with and with out silver are critically important. An in-depth review of silver's impact on filter performance is therefore important, however no such literature is available. This paper reviews filter mechanisms and efficacy for bacteria removal for cases with and without silver's addition. Method of silver application (dipping in a silver solution, painting, or co-firing) is assessed. Silver release and retention is also discussed. The findings from this paper illustrate that eluted silver contributes most to filter bacterial performance within the CWF receptacle. Silver application method, water quality and silver particle characteristics were also demonstrated to impact release. More research is required to understand silver's impact on a "slime layer" build-up on the internal filter surface. The

co-firing method of application results in the most consistent elution over time, at concentrations much lower than the alternatives. Finally, research into alternative metals to silver for enhanced disinfection present emerging opportunities within the CWF field with the potential of reducing cost without compromising the effectiveness of a CWF.

Keywords: Ceramic Water Filters, Filtration, Disinfection, Elution, Silver, International Development

2.1 Introduction

Access to safe water is not a universal challenge but is concentrated among the world's poorest and most vulnerable who are often disadvantaged by compounding economic, social, geographic, ethnic and gender-based marginalization (UNESCO 2015). Low-income communities are thus disproportionately burdened by water-related illness and child mortality rates. Furthermore, water-related illnesses can lead to a feedback loop where regular and/or chronic diarrhea can cause malnutrition and child stunting, which causes more diarrhea and hampers the child's long-term development, subsequently impairing socioeconomic development within that community (Heltberg 2009, K. Brown 2003). Therefore, as diarrhea is typically caused by a viral, parasitic or bacterial infection in the intestinal tract, which is spread through contact with food or water that has been contaminated by faecal matter (WHO 2017), increasing access to safe water is integral to mitigating this pervasive challenge (WHO; UNICEF 2014).

Methods of addressing such needs require alternative approaches than are typical in Western Nations though. Specifically, centralized treatment and distribution infrastructure have practical and economic challenges associated with short-term capital investment and long-term maintenance costs, as well as geographic limitations in the rural environment where safe water access is most limited. Therefore, implementation and use of decentralized, point-of-use water treatment solutions (POUWTS) to increase access to improved drinking water is in many cases, a more practical treatment option (WHO; UNICEF 2014, Sobsey, et al. 2008).

Many POUWTS, such as biosand filters, chemical disinfectants, natural and chemical coagulants, solar disinfection technologies (SoDIS), and others exist (see (Pagsuyoin, et al.

2015, Murphy, McBean and Farahbakhsh 2009, Santos, Pagsuyoin and Latayan 2016)), however this article is focused on ceramic water filters (CWF). Importantly, the objective of the work presented herein is not to proclaim the value of this technology over the others, as each possesses benefits and drawbacks which make them appropriate for different contexts. Rather, the intention is to review the literature for the CWF, for which there were approximately 700,000 CWFs estimated to be in use in 2014, serving 4 million people (van der Laan, et al. 2014). This review is therefore intended to facilitate and improve upon CWF applications to ensure its long-term robustness and sustainable use.

Ceramic Water Filters, which utilize a filtration mechanism to treat the water that passes through them, are a widely recommended POUWTS for implementation in Low- and Low-Middle Income contexts (WHO; UNICEF 2017). CWFs are easily manufacturable in the countries of intended use, as they are comprised primarily of a mixture of clay, firing material (FM; most commonly sawdust, rice husks or flour), water and often, a silver additive. The silver additive is the only component which may need to be imported. The wet mixture is typically pressed into a mold to form a pot shape, and after air drying, is heated to a high temperature in a kiln. During the heating process, the firing material burns away and leaves a porous network through which water may pass and subsequently be treated by size exclusion (van Halem, van der Laan and Heijman, et al. 2009, J. Rayner 2009).

The Ceramics Manufacturing Working Group (CMWG), a long-standing supporter of, and respected authority on CWFs, highlights silver as a key feature of the technology for the purpose of improving microbial disinfection performance and inhibiting bacterial growth along the interior filter barrier (CMWG 2010). Due, in part, to silver's high cost of approximately $3 USD per gram (Argenol Laboratories 2017), CWFs have been notably

more expensive than the financial means of its target market (Luoto, et al. 2012, Francis, et al. 2015, Burt, et al. 2017), indicating a thorough understanding of its disinfection contribution within a CWF context, as well as its mode of action, is imperative for the field.

This review paper focuses on the impact of silver as an additive to CWFs to highlight areas of interest for CWF manufacturers and researchers. The paper examines filter material ratio and particle size and discusses firing temperatures. In all areas, the paper attempts to highlight where further research is also needed. In particular, the paper addresses the state of understanding regarding microbial removal resulting from silver introduction into contaminated water, silver elution from a CWF into a filtrate receptacle during use, and how water quality influences both of those processes. A final section notes alternative emerging metal technologies with potential to progress the field.

2.2 Methods

Searches on SCOPUS and Web of Science were conducted independently for the three core themes reviewed in this article: Ceramic Water Filters, Silver Disinfection and Non-Silver Disinfection. For Ceramic Water Filters, the search string was ("Ceramic Filt*" OR "Ceramic Water Filt*" OR "CWF"). For Silver Disinfection, the search string was (Silver AND (disinfect* OR Bacteri*) AND Nanoparticle AND Water). For Non-Silver Disinfection, the string was ("Nanoparticle" AND (Disinfect* OR Bacteri*) AND Water). The results procured a total 1227, 3187 and 8068 documents, respectively, from the two databases combined; upon review the list was narrowed to the 134 references in this paper. These searches were originally conducted in Spring 2019, meaning articles published after this time were excluded. Some later papers were, however, included later in the writing process when authors felt appropriate. Moreover, titles were then reviewed for relevance, as

per the exclusion criteria for each category shown in Table 2.1. Any paper written in a language other than English was excluded from all categories, and duplicate papers were ignored during this review process.

2.3 Microbial Removal Mechanisms

As the primary aim of a POUWTS is to reduce the prevalence and impacts of diarrheal illnesses, the microbial removal efficiency of a CWF is of utmost importance. The removal of pathogenic microorganisms is mostly facilitated through filtration, which can be defined as "any process for the removal of solid particles from a suspension by passage of the suspension through a porous medium" (Crittenden, Trussell, et al. 2012a). Furthermore, it is well established that size-exclusion plays a fundamental role in CWF effectiveness (Panasewicz 2011, Bielefeldt, Kowalski and Summers 2009, Clark and Elmore 2011).

CMWG and other CWF manufacturing authorities also suggest silver significantly improves filter microbe removal efficacy (IDSSC n.d., Murray, Simpson and Fox 2011, van Halem, van der Laan and Heijman, et al. 2009, CMWG 2010). However, as will be demonstrated, the literature supporting the efficacy of the silver addition in both field and laboratory studies is quite variable and indicates that the understanding of how the silver acts with regards to disinfection requires more attention (Bielefeldt, Kowalski and Summers 2009, Kowalski 2008, J. Brown 2007, Lucier, Dickson-Anderson and Schuster-Wallace 2017). For instance, van der Laan et al. (2014) conducted a study to examine bacterial and virus removal from filters painted with colloidal silver, as well as ones without any added metals. The study results found that there was no statistically significant difference between filters with and without silver, as they yielded LRVs between 0.6 and 2.5 for filters with no silver and between 0.5 and 3.1 for filters with silver, when evaluating *E. coli*. The study also found

LRVs between 0.2 and 1 for filters without silver and between 0 and 1.4 for filters with silver, when evaluating MS2 Bacteriophages (van der Laan, et al. 2014). The removals were thus associated with the physical removal mechanisms of the CWF itself versus any disinfection action from the silver. An earlier study by Lantagne (2010) also found that when silver was painted on the CWF surface, coliform removals were similar to filters with no silver addition (Lantagne, et al. 2010). Finally, Rayner (2013) even found both significant and negligible changes to CWF microbe removal between disk filters with no silver, with 0.003 mg/g of silver, and 0.3 mg/g, depending on the clay and firing materials (Rayner, Zhang, et al. 2013). A comprehensive discussion of CWF microbe removal mechanisms may therefore assist in understanding why such discrepancies in data are being observed (see Section 2.3.2). This section reviews literature on CWF microbial removal behavior when absent of silver, followed by silver disinfection mechanisms and water quality impacts when acting independently in an aqueous environment.

2.3.1 Physical Microbe Removal

Size exclusion is the dominant mechanism for bacterial removal in a CWF, meaning filter performance is closely tied with pore size and porosity (Rayner, et al. 2017). Therefore, differences in the material parameters that lead to filter pore size and porosity also greatly impact filter performance. This section breaks down the bacterial removal in CWF relying on physical removal alone by examining the most commonly discussed CWF characteristics that impact microbe removal: material ratios, firing material particle size, filter thickness and final firing temperature.

From Tables 2.2 and 2.3, one may see that pots without silver addition have LRVs ranging from 0.4 to 8.17 (n=25), pots with silver have LRVs ranging from 0.5 to 6.7 (n=12), disks

without silver have LRVs ranging from 0.3 to 5.9 (n=36) and disks with silver have LRVs ranging from 2.2 to 6.3 (n=4). Thus, from a broad overview perspective it appears as though there is no significant difference between filters with and without silver. Disk filters also appear to act as representative samples for lab-scale experimentation. And finally, it appears that the variety of input material parameters used across the literature may create the observed ranges in filter performance; this concept is examined further in Sections 2.3.1.1 to 2.3.1.4.

2.3.1.1 Filtration Theory

CWFs operate through a combination of media layer surface removal (dominant) and depth filtration (secondary). Specifically, the majority of contaminants/microbes within the influent water are removed along the filter surface, as the majority are larger than the pore matrix throughout the ceramic filter body. However, removal also occurs through a combination of straining within the randomly oriented pathways throughout the filter matrix, and by getting caught within small "dead-end" pores. Thus, contaminants, are either blocked entry into the CWF body, or if able to enter, adhere to the ceramic pore walls or get trapped within pores that do not connect to the outer filter surface.

As contaminants are removed at the filter surface or trapped within the filter body, the water flowrate through the filter declines over time, while the contaminant removal increases. How much change is observed thus acts as a proxy for measuring how much of the contaminant load has been removed. Van Halem (2017) demonstrated that in high flowrate filters without silver (approx. 12-20 L/h initially), in both falling head filtration and continuous loading experiments, initial *E. coli* LRV values increased over filtration time as the filter became clogged with contaminants; specifically at initial water collection volumes of 60-85 L *E. coli*

LRV was <1, while at filter volumes of 240-320 L for the falling head filtration experiments and >1700 L for the continuous loading experiments, the *E. coli* LRV improved to approximately 2 in all cases. It was further noted that flowrates dropped upwards of 78% over the duration of filtered water collection, even with the inner surface of the filter having been scrubbed 5 times intermittently (van Halem, van der Laan and Soppe, et al. 2017). These results together illustrate that the vast majority of contaminants are either captured on the filter surface or trapped within the filter matrix itself and accumulate over time. The functioning of a CWF may thereby be defined by the physical properties and component parts that dictate water's ability to pass through the filter matrix.

Filter size influences flowrate and antibacterial performance by governing hydraulic head, and thus the pressure exerted by the water on the filters. Flow conditions through the pores, as well as the resulting shear force between the water and the pores, are therefore functions of the filter height, and by extension, size (Schweitzer, Cunningham and Mihelcic 2013, Annan, et al. 2014), among other parameters. CWF shape, however, has shown no significant impact on filter performance, as observed when comparing rounded- and frustum-bottomed filters, the most commonly utilized designs (Schweitzer, Cunningham and Mihelcic 2013). Clay minerology influences CWF performance by both dictating the proclivity of bacteria to attach to pore walls (Unuabonah, et al. 2018, Asadishad, Ghoshal and Tufenkji 2013), and by governing the amount of shrinkage experienced during the firing process (Oyanedel-Craver and Smith 2008). Further discussion of these parameters is, however, excluded from this article, as the impacts of optimizing it are believed to be superseded by the importance of using local materials/resources to the sustainability of the product (CMWG 2010).

Other manufacturing parameters are, however, easier to manipulate, and have been shown to closely relate with pore size and porosity (Yakub, et al. 2013, Rayner, et al. 2017). Specifically, the most commonly discussed CWF characteristics that impact microbe removal are material ratios, firing material particle size, filter thickness and final firing temperature, which are further discussed in this section.

2.3.1.2 Impact of Material Ratios

CWFs (without silver) are fabricated by first creating a homogeneous clay-FM mixture, where the clay is the ingredient which allows for the filter to be molded and subsequently maintain its shape after firing, and the FM burns away during the firing process to create a porous network throughout the clay matrix. Therefore, the ratio of these ingredients theoretically influences both bacterial removal and flowrate, as with more firing material added to the mixture, the more space is available for water, and consequently, bacteria to pass. A reduction in removal and an increase in filtration rate are thus anticipated. However, as may be observed in Tables 2.2 and 2.3, there is significant variability in the data within the literature, raising uncertainty in such theory.

Yakub et al. (2013) tested filter pots with the same sawdust particle size and material ratios of 35% and 50% (by volume). The study found that permeability increased, and tortuosity decreased as the percentage of sawdust content was increased, which corresponded to average Log Removal Values (LRV) of 6.36 ± 0.54 and 5.67 ± 2.50 for filters with 35% and 50% sawdust, respectively (Yakub, et al. 2013). The high level of variance for the filter with 50% sawdust, however, indicates that filters with this material ratio were more heterogeneous, making the data difficult to compare.

Rayner et al. (2017) conducted a comprehensive study on the impacts of firing material ratios, and their data was also inconclusive as to its impacts on bacteria removal. For example, disks with a sawdust particle size range of 0.250 mm – 0.595 mm and material ratios of 13.7%, 17% and 24% sawdust (by weight) yielded average LRV of 2.06 ± 1.33, 4.00 ± 0.285 and 2.78 ± 0.156 which shows no trend. Similarly variable results are found when the researchers tested filters with material ratios (by weight) of 18%, 19% and 25% milled rice husks of the same size range, yielding average LRVs of 1.93 ± 0.110, 1.26 ± 0.166 and 1.26 ± 0.097 (Rayner, et al. 2017). These findings illustrate that material ratios, regardless of the FM, are not controlling this filter performance metric. It is also noteworthy that variability decreased in the study conducted by Rayner et al. (2017), which sits contrary to the findings by Yakub et al. (2013). This point suggests compounding factors contribute to CWF bacteria removal by size exclusion.

In a third study, Soppe et al. (2015*)* reported no significant differences in LRV between filters with 24% and 31% rice husks (by weight), which yielded mean LRVs of 2.1 and 2.2, respectively (Soppe, et al. 2015). Interestingly, however, these researchers found weather influenced their results. For instance, the same filters made with 31% rice husks yielded an interquartile range for LRV of 3.6 – 3.9 in the dry season and 1.3 – 2.9 in the wet season, showing more variance and a lower average LRV during the wet season when compared with the dry. Thus, humidity or moisture content may need to be factored into the CWF production process.

Furthermore, the data presented herein suggests material ratios do not contribute to average LRV, and thus should not be considered a key parameter for this performance measure. Having said that, the same studies presented all demonstrate that there is indeed a linear

relationship between material ratios and flowrate (Soppe, et al. 2015, Rayner, et al. 2017), however analysis of this data lies beyond the scope of this review. These findings suggest that CWF flowrate may be increased without compromising microbe removal, though more research is necessary.

2.3.1.3 Impact of Firing Material Particle Size

From Tables 2.2 and 2.3, only 5 of 21 studies investigated FM particle size, even though it is considered a key design parameter by leading organizations in the field (J. Rayner 2009). As such, more research is certainly necessary in this domain to improve the field's understanding of this input parameter's impact.

Soppe et al. (2015) found a significant decrease in LRV with increasing particle size, with a median of 2.8 ± 0.7 (with an average flow rate of 3 L/h) for filters with <1mm FM size and 0.7 ±0.2 (with an average flowrate of 10.1 L/h) for filters with 0.5-1mm FM size (Soppe, et al. 2015). A less controlled particle range thus led to a higher LRV because smaller particles were included within the mixture, but it yielded larger variance. Conversely, the controlled, larger particle sizes had a smaller LRV with less variance, and a larger flowrate. Such results importantly indicate smaller particles may improve bacteria removal, but also highlight the challenges with reproducibility in the field.

Servi et al. (2013) also observed particle sizes of 388, 505 and 650 µm had very similar bacteria removal performances of 3.05 ± 0.8, 2.04 ± 0.5, and 2.77 ± 0.8, respectively, while a steep drop-off in LRV was observed after average size was increased to 780 µm and 925 µm, with average LRVs of 0.59 ± 0.2 and 0.83 ± 0.1, respectively (Servi, et al. 2013). Rayner et al. (2017), conversely, found that filters with 13.7% sawdust (by weight) and sawdust

particle size ranges of 0.250 – 0.595 mm, 0.595 – 1.19 mm and 1.19 – 2.38 mm yielded average LRVs of 2.06 ± 1.33, 4.43 ± 0.402 and 1.87 ± 0.261, respectively, showing no discernable relationship between FM size and LRV (Rayner, et al. 2017). Research from Varkey & Dlamini (2012) and Scannelle (2016) also found no discernable trend in LRV with changing FM size (see Tables 2.2 and 2.3, respectively) (Varkey and Dlamini 2012, Scannell 2016), demonstrating the need for more detailed analyses into its contribution to microbe removal by size exclusion.

2.3.1.4 Filter Thickness and Firing Temperature

Two other parameters that are considered to contribute to LRV are filter thickness and kiln firing temperature. Although multiple studies were found to discuss the importance of thickness as a design parameter (Yakub, et al. 2013, Schweitzer, Cunningham and Mihelcic 2013, CMWG 2010, Rayner, et al. 2017), only Servi et al. (2013*)* investigated its impact on LRV. The researchers found that LRV increased linearly from 0 to 0.6 as filter thickness increased from 3 to 20 mm with a particle size range of 500-1000 µm, and 0 to 2.1 when made with a particle size range of 400-500 µm (Servi, et al. 2013). The parameter interaction is logically consistent, as a thicker matrix provides more space for bacteria to be trapped within the pores, and smaller pores reduce space available for which the bacteria may pass, creating a compounding effect; a significant interaction between FM size and material ratios may also be expected as per the same reasoning, though such an investigation was not discovered in the literature.

Similarly, though many studies report the maximum firing temperature reached during filter fabrication (see Tables 2.2 and 2.3), Soppe et al. (2015*)* is the only study to investigate an impact of firing temperature on LRV. In their study, the researchers found the mean LRV of

filters fired at 800, 885 and 950 ^{0}C were 2.3, 2.1 and 1.9, respectively, demonstrating a slight decrease with increasing temperature (Soppe, et al. 2015). In addition, the mean pore diameters were reported to marginally increase from 27.8 µm to 28.9 µm to 30.6 µm with respective increases firing temperature (Soppe, et al. 2015). The interquartile LRV range (1.7-2.2, 1.5-2.6 and 1.1-2.8 with respective increased firing temperature) demonstrated an increase in data variability that also makes it difficult to confirm any significant difference between filters. Further to that end, the clay is vitrified at the peak temperature of the firing process, but sawdust burns away much earlier in the firing process; the final temperature reached is thus not the best measure of firing's influence on pore sizes, and consequently, LRV (CMWG 2010). Research into the firing process itself, and specifically the rate of temperature increase, is therefore needed to fully elucidate this design parameter's influence on filter performance.

2.3.2 Silver Disinfection

To understand the impact of silver on bactericidal/bacteriostatic effectiveness in a CWF system, one must first elucidate the mechanisms by which silver inhibits bacterial growth, or further causes cell lysis. This section reviews the disinfection mechanisms discussed in literature, followed by a discussion of the impacts that various nanoparticle and water quality characteristics have on bactericidal efficacy. Results on silver's disinfection efficacy from literature are also presented.

Silver disinfection is theorized to occur through two possible mechanisms, namely (1) Ag - Bacterial Interaction, and (2) Reactive Oxygen Species (ROS) generation (Le Ouay and Stellacci 2015, Lemire, Harrison and Turner 2013, Fauss, et al. 2014). Both mechanisms have demonstrated toxicity effects, as well as DNA replication inhibition (Duran, et al.

2016). However, the respective individual contributions or potential synergistic impacts are still under investigation.

2.3.2.1 Ag – Bacteria Interaction

One of the most commonly sourced mechanisms for silver-induced bactericide is the interaction between silver and the bacterial species, seen both in ionic (Ag^+) and nanoparticle (Ag-NP) forms. This interaction is hypothesized to result from electrostatic interactions and/or ionic bonding with sulphur or phosphate containing groups on the cell wall and within the membrane, resulting in bactericidal and bacteriostatic effects (Le Ouay and Stellacci 2015, Dror-Ehre, et al. 2009). However, there is a lack of consensus on their respective roles. Please note that bactericidal effects are defined here as effects which entirely kill the bacteria cell, whereas bacteriostatic effects are those which inhibit a cells ability to replicate DNA.

2.3.2.1.1 Silver and Cell Attachment

In terms of ionic silver, studies by Yamanaka et al. (2005) and others suggested that a monovalent cation like Ag^+ attaches to the negatively charged cell walls of the bacterium by electrostatic attraction , which leads to cell death as per the mechanisms explained in Section 2.3.2.1.2 (Dror-Ehre, et al. 2009, Yamanaka, Hara and Kudo 2005, Stoimenov, et al. 2002). Other researchers, however, have reported negatively charged Ag-NPs attachment to the cell walls, suggesting electrostatic attraction is not necessarily the governing mechanism (Sondi and Salopek-Sondi 2004, Morones, et al. 2005, Yakub and Soboyejo 2012). Rather, attachment of this nature is generally attributed to the bonding between silver nanoparticles and the sulphuric thiol groups on the cell membrane, which subsequently leads to toxicity effects or ROS generation and eventual cell death (Matsumura, et al. 2003, Shuang, et al.

2014). Therefore, it may be understood at the current juncture that electrostatic forces may play a role in silver-bacteria attachment, however silver bonding with sulphuric groups (primarily thiols (-SH)) on the cell wall is also central to initiating cell degradation or death (Shuang, et al. 2014).

2.3.2.1.2 Cell Lysis

After bonding has occurred and Ag^+ and/or Ag-NP accumulation develops along the cell wall, bacteriostatic and bactericidal processes ensue (Morones, et al. 2005, Feng, et al. 2000). Bacteriostasis results from a clustering of DNA in the centre of the cell known as the electron light region, which creates stress within the cell and inhibits its replication (Feng, et al. 2000). For *E. coli*, Ruparelia et al. (2008) found a minimum of 40-180 µg/mL of Ag-NPs (strain dependent) was required to inhibit growth of 99.9% of cells after 24 hours depending on size and morphology (Ruparelia, et al. 2008). Bactericidal processes occur upon greater accumulation of Ag^+ or Ag-NPs, which can both form pits in the cell wall, allowing for silver to infiltrate the cytoplasm and exhibit toxicity effects by bonding with DNA directly (Dror-Ehre, et al. 2009), deactivating cellular enzymes (Choi, et al. 2008) and releasing cytoplasm from within the cell, leading to degradation (El-Badawy, et al. 2011). For complete bactericide to occur in 99.9% of *E. coli* cells, Ruparelia et al. (2008) found a minimum concentration of 60-220 µg/mL of silver nanoparticles was required depending on the *E. coli* strain (Ruparelia, et al. 2008). Wu et al. (2018) found the same definitions of bacteriostasis and bactericide as outlined by Ruparelia et al. (2008) for *E. coli* required 10-60 µg/L and 20-140 µg/L, though the nanoparticles used by these researchers were of different size and morphology than Ruparelia et al. (2008), possibly explaining the observed differences (Wu, et al. 2018).

2.3.2.1.3 Reactive Oxygen Species (ROS) Generation

Although Ag-NP can contribute directly to bactericidal impacts, they can also react with oxygen in the water and release Reactive Oxygen species (ROS) as well as silver ions, as shown in Reaction 2.1 (Fauss, et al. 2014, Duran, et al. 2016, Lemire, Harrison and Turner 2013). Both silver ions and ROS also contribute to disinfection efficacy, though the relative contribution of Ag-NP, Ag+ and ROS is still unclear.

$$AgNP + O_2 \rightarrow Ag^+ + {}^{\bullet}O_2^- \qquad \text{[Reaction 2.1]}$$

In this case, ROS generation occurs before the nanoparticles interact with the cell, and thus ROS-based bacterial toxicity of this variety is an indirect result of silver presence. Other ROS, however, may be generated via electrochemical interactions with silver (McEvoy and Zhang 2014, Slavin, et al. 2017). For example, under photocatalytic conditions, the influx of energy from the light displaces one or more electrons from the valence band, which are subsequently pushed to the conductor band below (i.e. closer to the molecule). Resulting from this change, a positive hole (h^+) is created in the valence band and a free electron (e^-) in the conductor band, which makes the molecule subject to interactions with the aquatic environment as per Reactions 2.2 – 2.6 (Sirelkhatim, et al. 2015, McEvoy and Zhang 2014, Padmavathy and Vijayaraghavan 2008):

$$AgNP + hv \rightarrow h^+ + e^- \qquad \text{[Reaction 2.2]}$$

$$h^+ + H_2O \rightarrow {}^{\bullet}OH + H^+ \qquad \text{[Reaction 2.3]}$$

$$e^- + O_2 \rightarrow {}^{\bullet}O_2^- \qquad \text{[Reaction 2.4]}$$

$${}^{\bullet}O_2^- + H^+ \rightarrow HO_2^{\bullet} \qquad \text{[Reaction 2.5]}$$

$$HO_2^{\bullet} + H^+ + e^- \rightarrow H_2O_2$$ [Reaction 2.6]

Interestingly, these reactions have been shown to occur under dark conditions as well, however the instigation of the electron transfer and subsequent generation of ROS is still not well understood due to the fast rate at which the reactions occur (Fauss, et al. 2014). Having said that, both gram-positive and gram-negative bacteria have been shown to export electrons through their membrane when in contact with metals, suggesting the nanoparticles may be reduced by electrons on the bacterial surface, as per Reaction 2.7 (Ehrlich 2008):

$$AgNP + e^-(membrane) \rightarrow h^+ + e^-$$ [Reaction 2.7]

This particle-cell reaction is believed to occur because of silver bonding with sulfuric thiol groups and phosphoric DNA, which act as electron donors to initiate the reaction series of Reactions 2.3 – 2.6 (Choi, et al. 2008, Matsumura, et al. 2003, Kashida, et al. 2003). The kinetics with which silver and thiols/DNA react to form ROS has not been entirely elucidated, however Shuang et al. (2014) posit that ROS result as intermediates in the process of ionic silver bonding with thiols, eventually becoming Ag_2S (Shuang, et al. 2014). Silver-thiol bonding can lead to disinfection resulting from both ROS (Shuang, et al. 2014) and silver directly (Le Ouay and Stellacci 2015), although it is still unclear which mechanism dominates (Fauss, et al. 2014).

Furthermore, it has been observed that the small size and unstable nature of ROS allows them to penetrate the bacterial cell wall and enter the cytoplasm, accumulating inside and destroying DNA and stopping it from replicating cells from within (Choi, et al. 2008). Carlson et al. (2008) found that Ag-NPs in concentrations of 10, 25 and 50 µg/mL generated an increase in ROS concentration, measured as fluorescence intensity fold increase (unitless),

from 4 to 7 to 15, respectively, after 24 hours of incubation, which led to approximately 38, 65 and 80% reductions in cell viability, respectively (Carlson, et al. 2008). Park et al. (2009) even found that 40 and 60 minutes after $AgNO_3$ was added to water at 0.5 mg/L, a total LRV of 1.4 and 2.2 was achieved, respectively, to which ROS contributed 78% and 77% of the removal relative to Ag^+, respectively. Further, at a concentration of 1 mg/L, ROS contributed to 61% and 42% of total LRVs of 2.3 and 3.3 after 40 and 60 minutes, respectively. These findings suggest that concentration impacts the mechanics of disinfection (Park, et al. 2009).

2.3.2.2 Impacts of Water Quality Parameters

As is expected for any water treatment application, water quality parameters play a fundamental role in the efficacy of a given system. With CWFs in particular, researchers must be conscious of water quality to ensure the results will translate from laboratory waters into the field. This section therefore reviews the impact of pH, dissolved oxygen content (DO), charged species, and organic matter on silver disinfection.

2.3.2.2.1 pH Effect

The pH of water has a variety of effects on Ag-NP bactericide and/or bacteriostasis. Specifically, Ag^+ and ROS generation increase as pH decreases (Fauss, et al. 2014, He, Garg and Waite 2012, Fabrega, et al. 2009), which leads to a subsequent improvement in the silver's bacterial disinfection efficacy. In terms of ion generation, Liu & Hurt (2010) demonstrated that ion release results primarily from Reaction 2.8 (Liu and Hurt 2010), which indicates that with a lower pH (4-6), greater Ag^+ generation is observed than at more neutral pH (7-9) values. Specifically, as may be observed in Table 2.4, Ag^+ release increases from approximately 0.05 to 0.6 mg/L when pH decreases from pH of 9 to 4 (Liu and Hurt 2010).

ROS was not measured within this study, and thus its rate of generation under their study conditions is unknown.

Further, the impact of pH on Ag^+ release is hypothesized to improve bactericide as per two mechanisms. First, as the Ag-NP becomes oxidized, the oxide layer surrounding the nanoparticle becomes more soluble, thus making Ag^+ more available for disinfection. Second, nanoparticles are often stabilized with negatively charged capping agents, which when undissolved, aggregate Ag^+ ions. The resulting positively charged surface is consequently more attracted towards bacteria, improving disinfection (Lok, et al. 2007).

$$2AgNP_{(s)} + \frac{1}{2}O_{2(aq)} + 2H^+_{(aq)} \rightarrow 2Ag^+_{(aq)} + H_2O_{(l)} \qquad \text{[Reaction 2.8]}$$

At the current juncture, no studies have been found to date that detail the mechanisms behind the increase in ROS generation in the lower end of the environmental pH range (i.e. pH of 4-6). However, it is believed that this improvement results from an increase in Ag-NP oxidation, similar to the increase in Ag^+ generation (Fauss, et al. 2014). Moreover, as water chemistry, and particularly pH, inevitably differs across geographies and with changes in climate, it is important for CWF researchers and practitioners to understand how and why such changes may influence filter performance.

2.3.2.2.2 Dissolved Oxygen Content

Dissolved Oxygen (DO) has been proven to be a critical parameter in the disinfection properties of Ag-NP. Table 2.4, adapted from Liu & Hurt (2010), shows how the researchers demonstrated that the Ag^+ concentration in water was nearly ten times greater in natural water than "deoxygenated" water at a pH of 4 (Liu and Hurt 2010). Further, their study found almost no Ag^+ generation in "deoxygenated" water, even after 24 hours of incubation (Liu

and Hurt 2010). Fauss et al. (2014) also found that both Ag^+ and ROS generation dropped to zero after their test water was deoxygenated with a nitrogen purge, further reinforcing the necessity of DO in Ag-NP bactericidal effectiveness (Fauss, et al. 2014). The importance of oxygen in disinfection is understood from Reactions 2.1 and 2.8, where Ag-NP reacts directly with oxygen to release Ag^+ and/or ROS, which attack bacteria. More oxygen in the water therefore leads to faster conversion of nanoparticles into Ag^+ and/or ROS, increasing the concentration of these species and thereby improving the rate at which bacteria is killed (Xiu, Ma and Alvarez 2011).

In terms of application to CWFs, these observations are important because DO concentrations will vary depending on the water source. For example, heavily faecal-contaminated surface waters or groundwaters may have very low concentrations of DO, which could lead to a reduction in CWF effectiveness or ineffectiveness in terms of silver disinfection (Shwarzenbach, Gschwend and Imboden 2003). DO is also known to fluctuate with temperature and at different periods of the year, meaning CWF behavior may change depending on a certain geography. To date, no specific oxygen concentration has been established as the cut-off point when silver becomes ineffective, and no CWF studies have directly investigated the impacts of oxygen on removal performance, demonstrating a need for more research.

2.3.2.2.3 Charged Species

Research has demonstrated that divalent cations and halide ions can significantly impact silver bactericidal effectiveness, and are therefore the topics of discussion herein (Jin, et al. 2010, Bielefeldt, Stewart, et al. 2013).

Zhang et al. (2012) illustrated that water containing 1000 mg/L of divalent cations (Mg^{2+} or Ca^{2+}) and 11.5 mg/L of Ag-NPs led to an ionic silver release of 12-17 µg/L, whereas water containing silver and monovalent cations (Na^{+}) in the same concentrations saw an ionic silver release of 22-25 µg/L. These conditions correspond with disinfection performances of 72-73% and 81-82% after 20 hours of incubation, respectively (Zhang, Smith and Oyanedel-Craver 2012). Jin et al. (2010) found similar results, with the cell viability for the gram-negative bacterial species *Pseudomonas Putida (p. putida)*, which has similar size and biological characteristics to *E. coli*, increasing from 45% to 90% once the cations were added to the water (Jin, et al. 2010). These reductions in effectiveness have been attributed to the aggregation of the positive ions on the surface of Ag-NPs, increasing the hydrodynamic diameter of the nanoparticles and reducing their ability to penetrate the cell wall of the bacterium (See Section 2.3.2.1.2) (Jin, et al. 2010, Zhang, Smith and Oyanedel-Craver 2012). Thus, it is important to recognize that bacterial disinfection may be reduced in groundwaters or other waters with high divalent ions concentrations.

Chloride (Cl^{-}) is the most commonly discussed anionic species due to its high prevalence in nature and its common presence in water resulting from the use of chlorine as a disinfectant in municipal-scale water treatment facilities (Bielefeldt, Stewart, et al. 2013, Crittenden, Trussell, et al. 2012b). As such, Cl^{-} may be expected in influent water for a CWF if one is used in an urban environment or receives groundwater (Crittenden, Trussell, et al. 2012b). Moreover, due to the high affinity of silver nanoparticles and silver ions to Cl^{-} ($pK_{sp} = 9.75$) (Jin, et al. 2010), AgCl forms in an aqueous environment quite quickly. Choi et al. (2008) found this reaction and the formation of AgCl decreased the silver's disinfection effectiveness, as AgCl colloids inhibited *E. coli* growth by 66% compared with 100% by Ag^{+}

when added to water in a concentration of 4.2 µg/L (Choi, et al. 2008). Similar results were also exhibited by Levard et al. (2013), who found molar Cl/Ag ratios of 535 resulted in solid precipitates of AgCl to form, which, after 30 hours, resulted in the proportion of silver within their sample to decreased to 2% from approximately 8% when Ag-NPs were released into DI water only. Interestingly however, when the Cl/Ag ratio was increased to 2675 and further, 26750, Ag^+ release exponentially increased to approximately 6.5% and 17% dissolved content, respectively. These results also correlate closely with bacteriostatic efficacy, as the authors showed complete bacterial growth inhibition after 24 hours with $2x10^{-3}$ mol/L of Ag^+, Ag-NPs and Ag-NPs added to a 0.5 M NaCl solution, and only approximately 10% and 5% inhibition when the same concentrations were added to 0.1 M and 0.01 M NaCl solutions, respectively (Levard, et al. 2013). The reductions in bactericidal efficacy are therefore understood to occur as a result of the amount of dissolved species that are able to form. In other words, when Cl^- concentration is less than 3 orders of magnitude greater than the Ag^+ concentration (Levard, et al. 2013), Ag^+ and Cl^- form AgCl and this compound aggregates onto Ag-NPs to form a solid precipitate which inhibits bactericide by increasing the particle size, reducing its affinity towards the cell wall, and reducing the amount of release of Ag^+ from the AgNP (Baalousha, et al. 2013, Liu and Hurt 2010). However, at much higher concentrations of Cl^-, solid AgCl no longer forms, but rather Ag^+ and Cl^- form dissolved $AgCl_2^-$ or $AgCl_4^{3-}$, which do not as significantly reduce disinfection efficacy (Levard, et al. 2013). Similar effects have been found with other monovalent anions like hydroxide (OH^-), however the impacts are not as significant as those of Cl^- (Jin, et al. 2010).

2.3.2.2.4 Organic Matter

Organic matter is very commonly found in surface waters and has demonstrated to have a profound influence on silver nanoparticle disinfection (Zhang, Smith and Oyanedel-Craver 2012). Fabrega et al. (2009) showed that at a pH of 9, 2000 ppb of Ag-NP led to 85% bacterial growth inhibition, however essentially no growth inhibition was observed after 10 mg/L of Suwanee River humic acid was added to the water (Fabrega, et al. 2009). The reduction in efficacy is understood to result from the Ag-NP being unable to release Ag^+, as was clearly demonstrated by Liu & Hurt (2010). The researchers showed that at the same concentration of humic acid used by Fabrega et al. (2009), Ag^+ release dropped by 50%, and it continued to decrease along an exponential trend as natural organic matter (NOM) increased to 50 mg/L, at which point it was enumerated at nearly 0 mg/L of Ag^+ (Liu and Hurt 2010). The reduction in ion release has been ascribed to NOM sorption onto the nanoparticle, which creates a physical barrier through which the ionic silver cannot pass (Fabrega, et al. 2009). No literature was discovered on the impact of organic matter on ROS generation, however the complete failure of bacterial growth inhibition resulting from the presence of humic acid in the feed water demonstrated by Fabrega et al. (2009) suggests similar effects likely occur (Fabrega, et al. 2009). This impact is of particular importance to the CWF field, as the technology is often implemented in rural and remote communities that rely on surface water sources for drinking. As such, NOM can be expected in water sources, potentially impacting filter effectiveness in terms of silver disinfection.

2.3.2.3 Ag-NP Size and Shape

Physical nanoparticle characteristics have also been shown to impact disinfection efficacy; specifically, their size and shape are demonstrably important (Duran, et al. 2016, Morones, et al. 2005), because of the vastly increased specific surface area (i.e. surface area to

mass/volume ratio) of particles sized on the nanometer scale, relative to those of larger size (Lok, et al. 2007, Rai, Yadav and Gade 2009). Furthermore, Lok et al. (2007) found that 9.2 nm and 62 nm Ag-NP achieved the same bactericidal effectiveness (values unreported) when in concentrations of 12 µg/L and 108 µg/L, respectively. The researchers explained that the larger specific surface area of the smaller nanoparticle provided greater space for Ag^+ to be chemisorbed, which increased the nanoparticles' affinity towards the cell and therefore its bactericidal and bacteriostatic effectiveness in a lower concentration than its larger-sized counterpart (Lok, et al. 2007). Additionally, Helmlinger et al. (2016) found a direct correlation between specific surface area and nanoparticle dissolution, where after approximately 100 hours, particles of different shapes/morphologies with specific surface areas of 0.234, 0.100, 0.040 and 0.038 nm^{-1} dissolved such that the silver ion concentration was, on average, 40%, 27%, 15% and 10% of the silver-ultrapure water solution, respectively (Helmlinger, et al. 2016). These findings indicate that smaller particles with greater surface areas produce and adsorb more Ag^+, compounding to improve disinfection (Helmlinger, et al. 2016, Lok, et al. 2007).

Carlson et al. (2008) also showed that in a concentration of 25 µg/L, fluorescence intensity increased as sizes decreased, with reported values of (measured as fold increases) 2, 3 and 7.5 for Ag-NPs sized 55 nm, 30 nm and 15 nm, respectively. Smaller particles are therefore more easily oxidized by constituents in the aqueous environment than larger ones, leading to greater reductions of oxygen and subsequently vast differences in ROS generation. (Carlson, et al. 2008).

Aside from size, nanoparticle morphology has also been shown to contribute significantly to disinfection efficacy. For example, Pal et al. (2015) showed that truncated-triangular

nanoparticles achieved complete bacterial growth inhibition with only 10 µg/100 mL of silver in the culture medium after 26 hours, while after the same time period, spherical particles of the same size in a concentration of 12 µg/100 mL achieved only 40% growth inhibition (Pal, Tak and Song 2015). This result is particularly relevant because Collargol nanoparticles produced and sold by Argenol Laboratories, the most commonly used AgNPs in CWF manufacturing and research, are primarily spherical in shape (Panasewicz 2011, Oyanedel-Craver and Smith 2008, J. Rayner 2009). Further research into differently shaped AgNPs may therefore offer an opportunity to reduce the number of particles needed per filter, ultimately reducing the cost per unit.

2.3.3 Combined Physical and Silver Disinfection for Bacterial Removal in CWF

Within CWF research, the traditional understanding of the silver's effectiveness has been based largely on studies such as Oyanedel-Craver and Smith (2008), where after 75-85 minutes of filtration, filters painted with, and submerged in, a 600 mg/L colloidal silver solution yielded LRVs of 6 and 6.5, respectively, whereas filters without any silver yielded LRVs of 4.6 and 5.5 (Oyanedel-Craver and Smith 2008). And because of findings like these, Potters-for-Peace (PfP; the largest organization in this domain) states that "Silver is applied to the filter to achieve two objectives: 1) to take advantage of the bactericidal quality of silver in the purification of water as it is filtered; and 2) to prevent the growth of the "slime layer" of bacteria that can form on the filter wall" (p. 91) (CMWG 2010). This section therefore addresses both of these assertations, beginning with the latter.

2.3.3.1 Silver Impacts on "Slime Layer" Growth

Larimer's (2013) PhD thesis was the only document found to directly investigate this phenomenon, even though it is touted as one of two primary reasons to add silver to a filter (Larimer 2013, CMWG 2010). In it, the author shows that only 0.03% and 0.003% of *Mycobacterium smegmatis* remained on microporous track etched polycarbonate membranes (used to simulate a CWF surface) that were coated with an Ag-NP solution of 1.1 or 2.2 g/L, respectively, whereas the species grew by 1000% when no silver was deposited. Similarly, he found that 84% and 60% of *Mycobacterium Avium* remained on the same membranes when 1.1 and 2.2 g/L of Ag-NP solution was deposited, respectively, whereas the species grew by 637% without any silver presence (Larimer 2013). The silver concentrations selected, however, are orders of magnitude greater than what is typically added to a CWF. Additionally, Ag-NPs have been shown to disinfect gram-positive Actinobacteria (the phylum of the Mycobacteria genus) more than gram-negative Enterobacteriaceae (the phylum of the Escherichia genus) like *E. coli* (Yoon, et al. 2007), which makes these results difficult to generalize. This point is of particular importance as well, as diseases like cholera and typhoid are quite common causes of diarrhea in rural LIC and LMIC communities, which are gram-negative bacterial species. Thus, additional research that considers gram-negative bacterial species and at silver concentration more reflective of field conditions is necessary.

2.3.3.2 Silver Impacts during the Filtering Process

With filters, on average, flowing at a rate of approximately 1 – 4 L/hr through, on average, about 2.5 cm (1 inch) of filter material, the actual contact time between any single silver nanoparticle and bacterium is very brief. However, previous research (see Section 2.3.2.1) clearly indicates that silver disinfection is a somewhat slow process that requires time upwards of hours to inhibit DNA replication or exhibit toxicity effects (Pal, Tak and Song

2015, Yoon, et al. 2007, Rai, Yadav and Gade 2009). Therefore, the generalized hypothesis that that silver disinfects during filtration is questionable. Rather, it seems like a more likely that silver disinfects bacteria in the receptacle after filtration has already occurred.

Furthermore, Oyanedel-Craver and Smith (2008) reported initial differences in LRV between filters with and without silver of approximately 0.8. However, after about 20 minutes of filtration, the LRV difference between filter types became more prevalent, increasing to approximately 2 after 84 minutes (Oyanedel-Craver and Smith 2008). Figure S5 in their Supplementary Materials shows an initial flush of 0.5 mg/L of silver in the effluent after 20 minutes, which explains why the growing difference in LRV is observed (Oyanedel-Craver and Smith 2008). That is, impregnated filters introduced a lot of silver into the effluent initially, which reduced the bacterial concentration over time through disinfection action. Comparatively, the filter with no metal maintained a relatively constant effluent bacterial concentration, leading to an increasing difference between the two types of filters with time.

Abebe et al. (2015) reached a similar conundrum when attempting to remove *Cryptosporidium* from a variety of source waters – though silver was shown to effectively deactivate the oocysts in a batch reactor system, as observed through High-Resolution Differential Interference Contrast Imagery, the researchers were not able to identify whether it was filtration or disinfection that was responsible for their removal after passage through a ceramic disk in a separate experiment (Abebe, et al. 2015).

Direct evidence demonstrating the prevalence of post-filtration disinfection has also been reported by van der Laan (2014). The researchers found that *E. coli* LRV values taken less than 5 minutes after filtration were approximately 0.75 ± 0.25 for filters with no silver, 1.10 ± 0.4 with silver painted on the outside and 1.15 ± 0.5 for filters painted with silver both on

the inside and outside, showing no significant difference from silver addition. Conversely, filters painted with silver on both sides (in a separate experiment as above) yielded LRVs of 1.3 ± 0.7 after less than 5 minutes of storage compared with 3.8 ± 1.0 after 660 minutes of storage (van der Laan, et al. 2014).

2.4 Silver Elution

Silver elution is a significant feature of CWF performance, as it is necessary to ensure the concentration in the effluent remains below the WHO's recommended limit of 0.1 mg/L (WHO 2011). Daily ingestion of silver above this concentration may lead a lifetime accumulation greater than 10 g (WHO 2011), which can lead to negative health outcomes such as Argyria or even DNA damage (Fewtrell, Majuru and Hunter 2017). Guidelines developed by The Canadian Council of Ministers of the Environment (CCME) also stipulate total silver directly released into freshwater and marine environments should be in quantities no greater than 0.25 and 7.5 µg/L, respectively, to ensure the protection of aquatic life (CCME 2015). Conversely, as indicated in Section 2.3, silver elution also helps ensure sufficient microbe disinfection (van der Laan, et al. 2014). The release of ions and nanoparticles into the effluent initiates the interaction with bacteria that pass through the ceramic matrix, which improves removal with time (van der Laan, et al. 2014). Therefore, it is important to illuminate how silver attaches to, and is released from, ceramic to control for its impact in CWF performance.

Silver elution is a function of the particle characteristics, water chemistry and application method of the silver. Furthermore, subsequent disinfection is additionally impacted by whether particulate or dissolved silver is eluted. Thus, it is important to differentiate between particle release and ion release, as they occur via different mechanisms and the different

forms of silver will contribute to disinfection differently. As is seen herein however, a significantly greater quantity of research is required to better understand the mechanisms by which these processes occur, as limited studies have investigated elution (see Table 2.5). This section discusses application method, particle characteristics and water chemistry impacts on elution, and how each may contribute to disinfection.

2.4.1 Impacts of Application Method

While the addition of silver to CWF via painting and submerging methods are the most widespread in local manufacturing facilities (J. Rayner 2009, CMWG 2010), it appears, as seen in Table 2.5, that the co-firing (addition of a silver solution to the dry mixture before firing) leads to a more consistent and sustained release of silver into the effluent at quantities up to 1000 times less than the alternatives mentioned (Nunnelley, et al. 2016, Ehdaie, Rento, et al. 2017, Ren and Smith 2013).

Ren and Smith (2013) were the first to study this phenomenon, showing that over 180 minutes of filtration with synthetic water, filters impregnated with 2.76 mg/disk and 27.6 mg/disk released a consistent amount of total silver averaging 0.004 mg/L and 0.009 mg/L, respectively. When the influent water flowrate doubled, the disks impregnated with 2.76 mg/disk released between 0.003 - 0.007 mg/L, and those with 27.6 mg/disk released 0.015 mg/L consistently. Furthermore, after 360 minutes of filtration, the filters lost approximately 0.0045% and 0.001% of the initial silver content, respectively (Ren and Smith 2013). *Lyon-*Marion et al. (2018) found similar results for the co-firing method, where after 20 pore volumes of filtration with water containing an ionic strength (IS) of 10 mM ($NaNO_3$), co-fired filters released an average of 0.5% of the silver applied, compared with 11.8% and 8.7% for filters painted with $AgNO_3$ and Ag-NPs, respectively (Lyon-Marion, et al. 2018). These

results, as well as others, illustrate the comparatively low-level and consistent release of silver realized when filters are impregnated with the co-firing method (Lyon-Marion, et al. 2018, Nunnelley, et al. 2016, Ehdaie, Rento, et al. 2017, Kahler, et al. 2016, Ren and Smith 2013).

Conversely, studies that have investigated painting and dipping methods illustrate much higher releases, particularly at the beginning of filtration. For example, Ren and Smith (2013*)* show that disk filters painted and dipped to have an impregnated concentration of 2.76 mg/disk both initially released 11 mg/L of silver, decreasing to 0.4 mg/L and 1 mg/L, respectively, after 180 minutes of filtration. These values corresponded to a loss of approximately 1.20% and 1.25%of the initial silver content added for painted and submerged filters, respectively (Ren and Smith 2013). Mittelman et al. (2016) found agreeing results with disk filters painted with $AgNO_3$ initially releasing 1 mg/L, averaging just below 0.1 mg/L after about 50 pore volumes of filtration. Filters painted with Ag-NP had an initial release of approximately 0.5 mg/L, decreasing to 0.1 mg/L after 20 pore volumes, and then linearly decreasing to 0.03 mg/L after 160 pore volumes (Mittelman, et al. 2015). Lyon-Marion (2018) found almost identical results to those of Mittelman et al. (2016) (Lyon-Marion, et al. 2018). Furthermore, when scaled to full filters, Mikelonis et al. (2016) saw initial releases of 0.7 mg/L, 0.12 mg/L, 0.4 mg/L and 0.325 mg/L for filters painted with citrate-, polyvinylpyrollidine- (PVP), branched polyethylenimine- (BPEI) and Casein-stabilized nanoparticles, respectively, after exposure to a real surface water (Mikelonis, Lawler and Passalacqua 2016).

When taken together, all of these results, as well as the results presented in Table 2.5, highlight a trend where much more silver is lost when filters are painted with, or submerged

in, a silver solution, than when co-fired. These results align with the findings from Yakub and Soboyejo (2012), who enumerated the attachment force between silver and CWF material to be only 125 ± 32 nN after painting/submerging, which was attributed to van der Waals forces (Yakub and Soboyejo 2012). As such, the interfacial energy between the materials is low, which is why co-firing has exhibited more consistent release and less vulnerability to the impacts of water chemistry (Lyon-Marion, et al. 2018, Ren and Smith 2013). Because the energy between two interfaces (γ; i.e., silver and ceramic) is a function of enthalpy (H_s), temperature (T), and entropy (S_s), as shown in Equation 2.1 (Howe 1997). So, as temperature increases, the interfacial energy also decreases as per the second law of thermodynamics, explaining the stronger attachment of silver when filters are co-fired.

$$\gamma = H_s - TS_s \qquad [2.1]$$

2.4.2 Impacts of Silver Characteristics

Mittelman et al. (2015) demonstrated that between 0.008 - 1 mg/L of total silver from disk filters painted with $AgNO_3$ was released into the effluent, depending on variable water quality characteristics. Comparatively, disk filters painted with the same quantity of silver nanoparticles released between 0.006 – 0.4 mg/L of total silver under the same variable water quality conditions. With 10 mM of ionic strength background and after 10 pore volumes of filtration, $AgNO_3$-painted disks released 0.15 mg/L, whereas Ag-NP painted disks released 0.04 mg/L; after approximately 90 pore volumes of filtration though, the difference became negligible (Mittelman, et al. 2015). This observation may be due to the faster conversion of $AgNO_3$ into Ag^+, and the subsequent displacement of silver ions via cation exchange with ions in the influent, especially since painted silver is more vulnerable to removal (Mittelman,

et al. 2015). No other literature was found to evaluate the difference between Ag-NP and $AgNO_3$.

Mikelonis et al. (2016) demonstrated that negatively charged, electrostatically stabilized nanoparticles had greater attachment to an anodisc when positively charged than when negatively charged, whereas sterically stabilized nanoparticles exhibited the same amount of attachment to the anodisc, regardless of charge (Mikelonis, Youn and Lawler 2016). Having said that, Mikelonis et al. (2020) and Sullivan et al. (2017) illustrated that elution is impacted by stabilizing agent, as electrostatically stabilized Ag-NPs were eluted from ceramic surfaces in higher quantities than sterically stabilized ones, even if their charges were the same (Mikelonis, Rowles and Lawler 2020, Sullivan, Erickson and Oyandel-Craver 2017) The stabilizing agent of the silver nanoparticle was therefore found to impact elution from a CWF but not initial attachment. Electrostatically stabilized silver nanoparticles, meaning by ion associated adsorption onto the silver particle creating an electric charge that facilitates repulsion, are consequently more influenced by oppositely charged constituents in the surrounding environment. By contrast, sterically stabilized, meaning by polymer associated adsorption onto the silver particles that results in particle-particle repulsion, Ag-NP mobility is less influence by charged constituents in the environment. Surface functionalization is therefore closely related with application method, as certain particle stabilizations may yield more desirable elution levels if the CWF is painted or submerged, versus co-fired; further research into this relationship is highly recommended. Finally, Fauss et al. (2014) further reported improved disinfection with electrostatically stabilized Ag-NPs over steric stabilized particles (Fauss, et al. 2014), demonstrating the importance of considerations regarding consideration for what type of nanoparticle is utilized in a CWF for disinfection.

2.4.3 Impacts of Water Chemistry on Elution

Ionic strength was shown by Ren and Smith (2013) to improve silver retention when silver-spiked influent water (10 mg/L) was passed through a filter disk without silver impregnation for 100 minutes (6 pore volumes). The researchers found that with 50 nm diameter particles in the feed, 21%, 40% and 76% of the silver was retained when the water had a 1 mM, 10 mM and 50 mM IS, established using $MgSO_4$, respectively (pH not reported). More silver was also retained as the particle sizes increased, illustrating a compounding impact of ionic strength and size on retention (Ren and Smith 2013).

Interestingly Mittelman et al. (2015) demonstrated opposing results when disks were painted with a 200 mg/L silver solution and flushed with clear water for 12-15 hours, where they released approximately 0.006 mg/L, 0.08 mg/L and 0.8 mg/L with an influent water IS of 1 mM, 10 mM and 50 mM $NaNO_3$, respectively (Mittelman, et al. 2015). Similar results were reported by Lyon-Marion et al. (2018), where approximately 0.1 mg/L and 0.25 mg/L of silver was released from disk filters exposed to 1 mM and 10 mM IS of $NaNO_3$, respectively (Lyon-Marion, et al. 2018). The reason for the difference in observations is uncertain (Lyon-Marion, et al. 2018, Mittelman, et al. 2015), however is most likely due to $MgSO_4$ releasing divalent cations into the water, which cannot exchange with the monovalent Ag^+, relative to $NaNO_3$ releasing monovalent cations into the water, which can engage in ion exchange processes (Crittenden, Trussell, et al. 2012c). It may also be because the former study injected ions and silver into the feed, whereas the silver was only in the disks in the latter. As shown by Huynh and Chen (2011), a higher ionic strength leads to greater conversion of Ag-NPs to Ag^+, and divalent cations increase the rate of aggregation regardless of nanoparticle charge (Huynh and Chen 2011). It is thus most likely that the silver

nanoparticles in the feed were quickly converted into Ag^+, which then aggregated to form neutral agglomerates that were larger in size than the original nanoparticles, explaining why less than 1% ionic silver was enumerated by the researchers (Ren and Smith 2013). These larger agglomerates were therefore more retained because of their size, which is why more of the larger nanoparticles were retained with higher background IS, but size was irrelevant at a lower IS.

Changes in pH consistently created notable changes in silver leaching as well, however the nature of this impact is not completely understood. One explanation is that higher concentrations of hydrogen ions (e.g., pH = 5) creates a greater potential for cation exchange and thus displaces a greater quantity of silver, in comparison to higher pH levels (e.g., pH = 9) (Mittelman, et al. 2015, Bielefeldt, Stewart, et al. 2013). An alternative perspective however is that a step-wise reaction occurs, as shown in Reaction 2.8, whereby silver solid particles bond with oxygen in the water or on the ceramic surface, forming AgO or Ag_2O, which subsequently reacts with hydrogen and dissociates into dissolved silver ions and water (Hong, Smith and Srolovitz 1995, Nguyen, et al. 2014, Bielefeldt, Stewart, et al. 2013, Fauss, et al. 2014, Liu and Hurt 2010). Consequently, Mittelman et al. (2016), reported silver release to decrease from 0.6 mg/L to 0.1 mg/L and then 0.008 mg/L as pH increased from 5 to 7 to 9 sequentially (Mittelman, et al. 2015), demonstrating a clear relationship.

Chlorine concentration has a similar effect as well, as chlorine in the influent water can lead to silver chloride (AgCl) compounds forming as a precipitate (under conditions explained in Section 2.3.2.2.3), which detach the silver from the ceramic and can limit disinfection in the receptacle (Ouay and Stellacci 2015, Huynh and Chen 2011, Baalousha, et al. 2013, Bielefeldt, Stewart, et al. 2013). For example, Lyon-Marion et al. (2018) found essentially

no impact of 2 mg/L Cl_2 on elution of painted disks over 36 pore volumes, however they did find that when the influent concentration was subsequently increased to 4 mg/L of Cl_2, a decrease in elution from 0.1 mg/L to 0.06 mg/L was observed over 24 pore volumes (Lyon-Marion, et al. 2018).This effect is likely due to the retention of AgCl within the CWF matrix. No literature was discovered that discusses the influence of other monovalent anions (such as fluoride) on silver elution, which highlights an important research gap. Fluoride is, however, of concern, as it is abundant in natural waters, particularly in areas where water is exposed to volcanic rock and soil (Crittenden, Trussell, et al. 2012b). Unlike AgCl, however, AgF does not precipitate (Salt Lake Metals 2017), meaning it is unlikely to be retained within the CWF matrix, and thus more likely to appear in the effluent. As limited literature investigating AgF bactericidal efficacy is available, the impact of fluoride on subsequent disinfection potential remains uncertain. Further research into such effects would therefore be beneficial to the field.

2.5 Other Disinfection Enhancing Additives

While silver is the most common metal additive to CWFs for disinfection control, research on other metal species has also demonstrated some promise; most notably titanium dioxide (TiO_2), cupric oxide (CuO), and zinc oxide (ZnO) (Grass, Rensing and Solioz 2011, Foster, et al. 2011, Dimapilis, et al. 2018).

Like silver, these other metal species are suspected to release ions or ROS as mechanisms for disinfection, however TiO2 seldom releases Ti2+ and is only efficient as a source of ROS generation after photocatalysis (Foster, et al. 2011), so it is excluded from this discussion. CuO has demonstrated good levels of microbe removal as both ionic copper and as a source of ROS generation (Pandey, et al. 2012), and has also shown promise within a ceramic

system (Varkey and Dlamini 2012, Drelich, et al. 2017, Yakub and Soboyejo 2012, Ehdaie, Su, et al. 2020). Copper also has a higher recommended ingestion level of 2 mg/L (10 mg/day) which thus has lower longer term health risks than silver (WHO 2011). CCME, however, stipulates a maximum of 4 μg/L of total copper should be released directly into a receiving waterbody to ensure the protection of aquatic life (CCME 1987), which poses some environmental risk associated with its inclusion in a CWF system. This quantity is, however, significantly higher than the absolute minimum allowable concentration for silver release (see Section 2.4), suggesting its imposed risk is indeed lower than the current methods employed in the field.

ZnO has also shown to achieve bactericide under both dark and light conditions (Sirelkhatim, et al. 2015) and there is no WHO recommended limit for consumption of zinc, unlike the silver limit of 0.1 mg/L (WHO 2011). Zinc has even demonstrated to substantially reduce the incidence and severity of diarrheal episodes when ingested (Gitanjali and Weerasuriya 2011, Malik, et al. 2013) making it an attractive option to CWF application. CCME, however, states total zinc concentrations released directly into a waterbody with aquatic life should not exceed 9.1 μg/L, though that concentration may increase as water hardness, pH and dissolved organic carbon concentrations increase beyond 13.8 mg/L, 6.5 and 0.3 mg/L, respectively (CCME 2018). Furthermore, though environmental risks associated with zinc do indeed exist, they are significantly lower than those posed by the inclusion of silver or copper in a CWF.

No literature was discovered that discusses a ZnO composite CWF; only an unpublished study by van Halem (2006) showed it was retained within a filter (J. Rayner 2009). The value of using ZnO as a replacement or supplement for AgNPs is therefore unexplored.

2.5.1 CuO Disinfection

CuO disinfection has been a recognized phenomenon dating back to ancient civilizations using copper vessels to ensure safe storage of water (Grass, Rensing and Solioz 2011). Following a similar mechanistic pathway as both silver and zinc, CuO has shown to generate ROS via Reaction 2.9 or 2.10, as well as release copper ions, which also have bactericidal impacts (Grass, Rensing and Solioz 2011, Thurman and Gerba 1989). Though far less studied within the CWF research field, CuO has demonstrated comparable, and sometimes even greater, bactericidal effectiveness than AgNP, highlighting an area of great potential for manufacturing adjustments. For example, Yoon et al. (2007*)* found 33.5 µg/mL (0.421 µmol/mL) of CuO nanoparticles were required to achieve an E.coli LRV of 1 after 24 hours of incubation compared with 58.4 µg/mL (0.541 µmol/mL) of Ag-NPs (Yoon, et al. 2007). Conversely, Ananth et al. (2015*)* found the minimum inhibitory concentration for *E. coli* after 12 hours was 6.25 µg/mL (0.079 µmol/mL) for CuO compared with 1.56 µg/mL (0.014 µmol/mL) of Ag, illustrating opposite results (Ananth, et al. 2015). Pandey et al. (2012) found LRVs of 2.5 and 4.3 with CuO concentrations of 10 µg/mL (0.126 µmol/mL) and 25 µg/mL (0.314 µmol/mL), respectively, against *E. coli*, however they did not compare effects with silver (Pandey, et al. 2012).

$$Cu^{+} + H_2O_2 \rightarrow Cu^{2+} + OH^{-} + OH^{\bullet} \qquad \text{[Reaction 2.9]}$$

$$2Cu^{+} + 2H^{+} + O_2 \rightarrow 2Cu^{2+} + H_2O_2 \qquad \text{[Reaction 2.10]}$$

In terms of application to the CWF field, only three studies were found within the literature that specifically studied copper addition to a CWF, which were Varkey et al. (2012), Lucier et al. (2017) and Jackson, Smith and Edokpayi (2019). The lattermost researchers found that

filters co-fired with 2 and 4 g of $Cu(NO_3)_2$ both yielded LRVs of 3.54 and released 3.5 and 9.5 µg/L of copper, respectively, whereas filters painted with 0.4 g AgNP achieved an LRV of 3.76 and released 21.5 µg/L of silver (Jackson, Smith and Edokpayi 2019). Lucier et al. (2017) found that filters without any metals yielded interquartile LRV ranges of 3 to 6, whereas filters co-fired with CuO and Ag (concentrations not reported) yielded LRVs ranging from 3 to 6 and 3.2 to 6, respectively (Lucier, Dickson-Anderson and Schuster-Wallace 2017). The former results indicate better disinfection for AgNP-impregnated filters, whereas the latter illustrates no discernable difference between any of the filters evaluated. Varkey et al. (2012) reported a marginal difference in bacteria removal when adding a copper mesh directly into the receiving receptacle and no copper in the CWF itself; Removals were reported as 100% and 99.4% removal of *E. coli* with and without the copper mesh, respectively (Varkey and Dlamini 2012). Additional research on copper disinfection within a CWF system would benefit the field, as this alternative metal may offer CWF manufacturers more diversity in their supply chain so there does not have to be such a great reliance on silver for enhanced disinfection.

2.5.2 ZnO Disinfection

As discussed, ZnO nanoparticles (ZnO-NP) disinfect bacteria with similar mechanisms to silver and copper, where zinc ions (Zn^{2+}) and/or ROS inhibit DNA replication within the cell organism or exhibit toxic effects (Yoon, et al. 2007, Dimapilis, et al. 2018). Most literature on this compound cites UV and visible light activation as the source of ROS generation via electron-hole pairs, as shown through Reaction 2.11 and Reaction 2.3 – 2.6. For example, Padmavathy and Vijayaraghavan (2008) found that under these conditions, ZnO had a 90% bactericidal efficiency after 24 hours of incubation, which was reportedly due to ROS

generation on the nanoparticle surface (Padmavathy and Vijayaraghavan 2008). Hirota et al. (2010) and others, however, have found ROS generated under dark conditions as well (Adams, Lyon and Alvarez 2006, Sirelkhatim, et al. 2015, Hirota, et al. 2010). Song et al. (2010) found under these conditions, the nanoparticle interacts with the cell and causes damage to the mitochondria, which releases oxygen that then quickly reacts to become ROS and improve disinfection efficacy over time (Song, et al. 2010).

$$ZnO + hv \rightarrow e^- + h^+ \quad \text{[Reaction 2.11]}$$

$$h^+ + H_2O \rightarrow OH^\bullet + H^+ \quad \text{[Reaction 2.3]}$$

$$e^- + O_2 \rightarrow O_2^{\bullet -} \quad \text{[Reaction 2.4]}$$

$$O_2^{\bullet -} + H^+ \rightarrow HO_2^\bullet \quad \text{[Reaction 2.5]}$$

$$HO_2^\bullet + H^+ \rightarrow H_2O_2 \quad \text{[Reaction 2.6]}$$

The same researchers, however, found that ROS was not as impactful as dissolved, ionic zinc (Zn^{2+}) in disinfection under dark conditions, as they found ionic zinc resulted in a 55% reduction in cell viability, and a zinc oxide suspension resulted in a 65% reduction with "fine-ZnO" particles at a concentration of 100 µg/mL (Song, et al. 2010). Ionic zinc is believed to disinfect the bacterial cell by attaching to the cell outer membrane by either electrostatic forces or by bonding with the sulfuric thiol- or phosphoric protein groups, causing damage to the cellular wall and eventually infiltrating the cytoplasm, destroying the cell from within (Maret 2004, Qu, Alvarez and Li 2013). Contrary to those findings, Joe et al. (2017) found an 87% reduction in cell viability after 6 hours of exposure to 2.85 mg/L of ZnO-NPs with only 11% of that solution in ionic form. The researchers therefore concluded that Zn^{2+} was not in a high enough concentration to be toxic to the cell, and therefore could not control

bacterial disinfection. Rather, they hypothesize that ZnO-NPs attach to the cell membrane and initiate the bactericidal process, and in so doing, release Zn^{2+} onto the cell itself, which improves bactericide even further over time (Joe, et al. 2017). Regardless of which antibacterial pathway is dominant, the fact that disinfection is observed under both light and dark conditions is important to the CWF field, as filtrate is most often kept under dark conditions. And as discussed in Section 2.3.3, metal-based disinfection is predominantly active in the receptacle (i.e., after filtration), meaning those conditions are of particular importance to the field for further research. These findings further highlight the importance of studying the impact of ZnO within a CWF system, as no research has been discovered that studies zinc-impregnated filters.

2.6 Conclusion

Diarrheal illnesses claim the lives of hundreds of thousands of children each year, meaning access to safe drinking water is a key element of a global strategy to eradicate this challenge. Its disruptive impacts, though, are clearly observed along socio-economic lines, where those most significantly afflicted are most often low-income groups living in a rural setting. POUWTS are therefore widely regarded as useful technological solutions that may be implemented with immediacy in rural environments, Ceramic Water Filters being one of the most common.

Silver is typically added to a CWF as an antimicrobial agent to improve the bactericidal efficacy of the technology. However, the contribution of silver to the CWF has been, at times, unclear. Importantly, in this review of the literature, it was demonstrated that when evaluating filter bacteria removal immediately after filtration, those with silver do not perform significantly better than those without. Only after storage time do filters with silver

reduce the bacterial concentration significantly more than those without, illustrating that silver nanoparticles add to the overall filter performance primarily in the receptacle. Thus, user behavior will impact the level of safety from the CWF – does the user consume water immediately after filtration or wait a certain amount of time?

Furthermore, this review highlights that silver elution is thus critical to the realization of safe drinking water for the technology's users. Its relationship with disinfection kinetics in the receptacle, however, remains understudied, demonstrating an important research gap. Specific attention should be placed on co-fired filters, as results from literature suggest it is a superior silver application method to painting and submerging in terms of release consistency, concentration, and robustness of performance across diverse water qualities. Considerations of influent water quality, physical filter characteristics and silver type are also critical. Further research is also still required to elucidate the metallic influence on the formation of a microbial "slime" layer along the inner part of the filter, and how the filter performs over its expected lifespan.

Review of literature regarding alternative metals to silver for enhanced disinfection, such as Zinc Oxide (ZnO) and Cupric Oxide (CuO), also present emerging opportunities for new areas of research with the potential of reducing cost and increasing metal nanoparticle options without compromising the effectiveness of a CWF. Greater attention to this research area could expand the potential reach and effectiveness of this technology, importantly increasing access for more marginalized individuals and communities.

2.6.1 List of Tables

Table 2.1 Exclusion Criteria for Review

Ceramic Water Filters	**Silver Disinfection**	**Non-Silver Disinfection**
(1) Community User-based Field Studies	(1) Non-drinking-water-treatment applications	(1) Non-drinking-water-treatment applications
(2) Studies that do not evaluate bacteria removal as part of the study	(2) Non-nanoparticle silver	(2) Non-nanoparticles
	(3) Photocatalytic research	(3) Photocatalytic research
	(4) Studies that do not evaluate bacteria removal as part of the study	(4) Studies that do not evaluate bacteria removal as part of the study
		(5) Silver-related nanoparticle research

Table 2.2 Summary of CWF Pot Parameters and Performance Outcomes

Reference	Pot/Disk	Minimum LRV[1]	Maximum LRV	CWF[2]/ CWF + Ag[3]	%Firing Material	By Weight or Volume	Firing Material Sieve Size (<x)(mm)	Firing Material Type	Maximum Firing Temperature	Average Flowrate (L/h)
(Abebe, et al. 2015)	POT	2.4	4.3	CWF	50	vol[5]	0.595	SD[7]	NR	1.7
(Bielefeldt, Kowalski and Summers 2009)	POT	0.4	4.05	CWF	40	vol	1.2	SD	887	1.7-1.9
	POT	0.5	4.45*	CWF + Ag	40	vol	1.2	SD	887	0.8-1.8
(J. Brown 2007)	POT	2	2.6	CWF + Ag	NR[4]	NR	NR	RH[8]	870	2
	POT	1.5	2.6	CWF	NR	NR	NR	RH	870	2
(Clark and Elmore 2011)	POT	1.2	3.4	CWF	10	wt[6]	4.8	SD	NR	1.2
(Guerrero-Latorre, et al. 2015)	POT	0.68	2.9	CWF	23	wt	0.25	RH	950	2.25
(Lantagne, et al. 2010)	POT	3.1	6.1*	CWF + Ag	40	vol	NR	SD	NR	1.5
	POT	3	6	CWF + Ag	47	vol	0.0003	SD	NR	1.5
(Lucier, Dickson-Anderson and Schuster-Wallace 2017)	POT	2	6.25	CWF	47	vol	0.0003	SD	870	0.8385
	POT	3	6.68	CWF + Ag	47	vol	0.0003	SD	870	1.129
(Mikelonis, Lawler and Passalacqua 2016)	POT	1	1.5	CWF	20	wt	NR	RH	NR	3.6
	POT	1.9	4.5*	CWF + Ag	20	wt	NR	RH	NR	3.6
(Kowalski 2008)	POT	0.446	3.753*	CWF	40	vol	NR	SD	NR	0.6-0.8
	POT	0.461	5.183*	CWF + Ag	40	vol	NR	SD	NR	0.6-0.8
(Panasewicz 2011)	POT	2	2.3	CWF	NR	NR	NR	SD	NR	1.8
	POT	3.75	4	CWF + Ag	NR	NR	NR	SD	NR	2
	POT	1.95	2.95	CWF + Ag	24	wt	NR	RH	NR	1.4

Table 2.2 (cont.) Summary of CWF Pot Parameters and Performance Outcomes

Reference	Pot/Disk	Minimum LRV	Maximum LRV	CWF/ CWF + Ag	%Firing Material	By Weight or Volume	Firing Material Sieve Size (<x)(mm)	Firing Material Type	Maximum Firing Temperature	Average Flowrate (L/h)
(Soppe, et al. 2015)	POT	1.5	3.5	CWF	24	wt	0.5	RH	885	5.21
	POT	2.1	4.5	CWF	26	wt	0.5	RH	885	9.194
	POT	1.5	4	CWF	28	wt	0.5	RH	885	14.725
	POT	1.5	3.25	CWF	29	wt	0.5	RH	885	16.535
	POT	1.5	3.75	CWF	31	wt	0.5	RH	885	19.75
(van Halem, van der Laan and Soppe, et al. 2017)	POT	0.8	1	CWF	24	wt	0.5	RH	NR	3.5
	POT	0.75	2	CWF	28	wt	0.5	RH	NR	11.5
	POT	0.75	2.5	CWF	31	wt	0.5	RH	NR	19
	POT	1.1	1.75	CWF + Ag	24	wt	0.5	RH	NR	3.5
	POT	0.85	1.5	CWF + Ag	28	wt	0.5	RH	NR	11.5
	POT	0.85	1.75	CWF + Ag	31	wt	0.5	RH	NR	19
(Varkey and Dlamini 2012)	POT	NR	4*	CWF	50	NR	0.3	SD	850	NR
	POT	NR	4*	CWF	66	NR	0.3	SD	850	0.05
	POT	NR	4*	CWF	50	NR	0.6	SD	850	NR
	POT	NR	4*	CWF	66	NR	0.6	SD	850	0.11
	POT	NR	3.98	CWF	50	NR	0.9	SD	850	NR
	POT	NR	3.98	CWF	66	NR	0.9	SD	850	0.14
(Yakub, et al. 2013)	POT	3.17	8.17*	CWF	50	vol	0.00107	SD	955	1.65
	POT	5.82	6.9	CWF	35	vol	0.00051	SD	955	0.5

* = 100% of *E. coli* was removed; 1 LRV = Log Removal Value; 2 CWF = Ceramic Water Filter (no silver); 3 CWF + Ag = Ceramic Water Filter (with silver); 4 NR = Not Reported; 5 vol = By Volume; 6 wt = By Weight; 7 SD = Sawdust; 8 RH = Rice Husks;

Table 2.3 Summary of CWF Disk Parameters and Performance Outcomes

Reference	Pot/Disk	Minimum LRV[1]	Maximum LRV	CWF[2]/ CWF + Ag[3]	%Firing Material	By Weight or Volume	Firing Material Sieve Size (<x)(mm)	Firing Material Type	Maximum Firing Temperature	Average Flowrate (L/h)
(Rayner, Zhang, et al. 2013)	DISK	2.2	6.7*	CWF + Ag	15	wt	1.19	SD	550	0.03
(Rayner, et al. 2017)	DISK	1.55	2.7	CWF	14	wt[6]	0.6	SD[7]	NR[4]	0.02
	DISK	3.15	4.7	CWF	17	wt	0.6	SD	NR	0.02
	DISK	2.5	4.95	CWF	20	wt	0.6	SD	NR	0.03
	DISK	2.1	4.1	CWF	24	wt	0.6	SD	NR	0.21
	DISK	2.85	6.3	CWF	14	wt	1.19	SD	NR	0.03
	DISK	1.3	4.1	CWF	17	wt	1.19	SD	NR	0.1
	DISK	1.55	4.9	CWF	20	wt	1.19	SD	NR	0.125
	DISK	1	2.7	CWF	11	wt	2.38	SD	NR	0.07
	DISK	0.6	4.2	CWF	14	wt	2.38	SD	NR	0.0875
	DISK	1.3	2.4	CWF	18	wt	0.6	RH[8]	NR	0.19
	DISK	0.6	2.8	CWF	19	wt	0.6	RH	NR	0.195
	DISK	0.8	1.8	CWF	25	wt	0.6	RH	NR	0.378
	DISK	0.7	2	CWF	8	wt	1.19	RH	NR	0.055
	DISK	0.8	2.2	CWF	12	wt	1.19	RH	NR	0.055
	DISK	0.7	1.5	CWF	17	wt	1.19	RH	NR	0.295
	DISK	0.55	2.5	CWF	10	wt	2.38	RH	NR	0.4
	DISK	0.5	2.2	CWF	17	wt	2.38	RH	NR	0.64
	DISK	0.35	1.6	CWF	25	wt	2.38	RH	NR	1.25
(Nunnelley, et al. 2016)	DISK	4.8	5.9	CWF	NR	NR	0.841	SD	NR	NR
	DISK	4	5.6	CWF + Ag	NR	NR	0.841	SD	NR	NR
(Oyanedel-Craver and Smith 2008)	DISK	3.1	5.4	CWF	10	wt	NR	F[9]	900	1.17234
	DISK	5.3	6.3	CWF + Ag	10	wt	NR	F	900	1.17234

Table 2.3 (cont.): Summary of CWF Disk Parameters and Performance Outcomes

Reference	Pot/Disk	Minimum LRV[1]	Maximum LRV	CWF[2]/ CWF + Ag[3]	%Firing Material	By Weight or Volume	Firing Material Sieve Size (<x)(mm)	Firing Material Type	Maximum Firing Temperature	Average Flowrate (L/h)
(Scannell 2016)	DISK	3.25	4.1	CWF	NR[4]	NR	0.297	SD[7]	890	0.1785
	DISK	3.9	4.1	CWF	NR	NR	0.433	SD	890	0.132
	DISK	2	4.75	CWF	NR	NR	0.569	SD	890	0.1455
	DISK	2.75	3.6	CWF	NR	NR	0.705	SD	890	0.4275
	DISK	3.25	3.5	CWF	NR	NR	0.841	SD	890	0.309
	DISK	2.5	2.75	CWF	NR	NR	0.297	RH[8]	890	0.148
	DISK	1.5	2.5	CWF	NR	NR	0.433	RH	890	0.144
	DISK	1.5	2.75	CWF	NR	NR	0.569	RH	890	0.222
	DISK	1.9	2.25	CWF	NR	NR	0.705	RH	890	0.445
	DISK	1.6	2.5	CWF	NR	NR	0.841	RH	890	0.328
(Servi, et al. 2013)	DISK	2.25	3.98	CWF	20	wt[6]	0.42	RH	NR	0.0134
	DISK	1.6	2.6	CWF	20	wt	0.59	RH	NR	0.0123
	DISK	2.3	3.65	CWF	20	wt	0.71	RH	NR	0.0783
	DISK	0.4	0.8	CWF	20	wt	0.85	RH	NR	1.92
	DISK	0.75	0.9	CWF	20	wt	1	RH	NR	1.5
(Zhang and Oyanedel-Craver 2013)	DISK	NR	4.42	CWF + Ag	10	wt	NR	F[9]	900	0.222
	DISK	4.22	4.34	CWF	10	wt	NR	F	900	0.222

* = 100% of *E. coli* was removed; 1 LRV = Log Removal Value; 2 CWF = Ceramic Water Filter (no silver); 3 CWF + Ag = Ceramic Water Filter (with silver); 4 NR = Not Reported; 5 vol = By Volume; 6 wt = By Weight; 7 SD = Sawdust; 8 RH = Rice Husks; 9 F = Flour

Table 2.4 Dissolved Silver (Ag+) concentration resulting from Ag-NP conversion at various pH and dissolved oxygen levels. (Data adapted from Liu & Hurt (2010))

pH	Incubation Time (hours)	Dissolved Silver Concentration (mg/L)
Dissolved Oxygen Concentration: 9.1 mg/L		
9	24	0.03
8	24	0.085
7.4	24	0.12
5.68	24	0.29
5.6	24	0.315
5	24	0.39
4	24	0.58
5.68	3	0.055
5.68	6	0.115
5.68	12	0.17
Dissolved Oxygen Concentration: <0.1 mg/L		
5.68	3	0.005
5.68	6	0.002
5.68	12	0.002
5.68	24	0.002
4	24	0.05

Table 2.5 Summary of Elution Data from CWFs painted or co-fired with Silver Nanoparticles

Reference	Mass of Silver Applied	Silver Application Method	Elution (mg/L)	Comment
(Oyanedel-Craver and Smith 2008)	0.0028 -0.036 mg Ag/g Filter	Painted	0.025 - 0.5	Water quality not reported
(van der Laan, et al. 2014)	300 mL of 0.00215 M Solution	Painted	0.02 - 0.225	Natural surface water used for entire test
(Mikelonis, Lawler and Passalacqua 2016)	0.018 mg Ag/g Filter	Painted	0 - 0.8	Influent water was either natural dugout water or natural rainwater. 4 different nanoparticle stabilizing agents evaluated
(Mittelman, et al. 2015)	0.03 mg/g Filter	Painted	0.03 - 1	Influent water with 10 mM NaNO3. Filters painted with both AgNO3 and Ag-NP. Range is difference between initial and after 160 pore volumes of filtration
(Kowalski 2008)	300 mL of 0.00215 M Solution	Painted	0.004 - 0.09	Water quality not reported. Results for Potters for Peace filters reapplied with silver in 2003. Experimentation conducted on filters before present study.
(Ehdaie, Su, et al. 2020)	9.28 mg Ag/g Filter Tablet	Co-Fired	0.15	Water chemistry not reported. Co-fired filters are tablets submerged in water. Applied silver concentration extrapolated.
	92.8 mg Ag/g Filter Tablet	Co-Fired	5	
(Lyon-Marion, et al. 2018)	0.05 mg Ag/g	Co-Fired	0.01125	10 mM NaNO3 in synthetic feed water Concentration within first 20 Pore Volumes of filtration. More silver released from filters painted with AgNO3 than Ag-NP (painted).
	0.03 mg Ag/g	Painted	0.1175 - 0.1593	
(Ren and Smith 2013)	2.76 mg Ag/g Filter	Co-Fired	0.003 - 0.004	Synthethic Hard Water used in feed (Alkalinity 110-120 mg/L CaCO3) Elution slightly increases when influent flowrate increases.
	27.6 mg Ag/g Filter	Co-Fired	0.009 - 0.015	
	2.76 mg Ag/g Filter	Painted	0.008 - 11	Same influent water used as above. Elution decreases log-linearly, decreasing faster after influent flowrate increased.
	2.76 mg Ag/g Filter	Dipped	0.005 - 11	
	24.8 mg/g Filter	Co-Fired	0.0025 - 0.1	

(Jackson and Smith 2018)	49.6 mg/g Filter	Co-Fired	0.0025 - 0.045	Synthetic water tested with 10 mM phosphate buffer solution
	4.96 mg Ag/g Filter	Painted	0.03 - 0.20	
(Jackson, Smith and Edokpayi 2019)	0.1176 mg Ag/g Filter	Co-Fired	0.0005 - 0.0015	Natural groundwater used as influent. Effluent concentrations taken 14 hours after filtration. Filter weight assumed from mass of clay used
	0.588 mg Ag/g Filter	Co-Fired	0.0025 - 0.008	
	0.1176 mg Ag/g Filter	Painted	0.0115 - 0.028	
(Ehdaie, Rento, et al. 2017)	0.088 mg Ag/g Filter	Painted	0.007 - 0.046	Tested in natural water. Applied silver concentration extrapolated. Samples collected over 52 weeks in field study
	9.28 mg Ag/g Filter Tablet	Co-Fired	0.001 - 0.003	Tested in natural water. Data from tablets submerged in water. Samples collected over 52 weeks in field study
(Nunnelley, et al. 2016)	5.35 mg/filter	Painted	0.025 - 2	Influent water with 10 mM phosphate buffer solution. Filter mass not reported. Silver concentration decreases exponentially with time
	26.75mg/filter	Co-Fired	0.002 - 0.2	
	53.5 mg/filter	Co-Fired	0.002 - 0.03	

Chapter 3: Field-Focused Literature Review

SDG 6.1 proposes that all individuals worldwide should have access to safely managed drinking water services (SMDWS) by 2030. The UNICEF/WHO Joint Monitoring Program (JMP) defines a SMDWS as "an improved source that is accessible on premises, available when needed, and free from faecal and priority chemical contamination"; an improved source is that which is not directly collected as surface or rainwater (UN 2015). References to *who* is managing these services, what the *management* of a service entails, and what constitutes a *service* in general, are importantly absent (JMP 2021). A multiplicity of technical and administrative drinking water source arrangements may therefore be considered a SMDWS (Bain, et al. 2021). Significant research towards identifying optimum methods for accelerating associated progress has responsively emerged (Milman, Kumpel and Lane 2021, Martin, et al. 2018, Ginja, Gallagher and Keenan 2021, Clasen, et al. 2015).

An approach that has received notable attention is the implementation of point of use water treatment solutions (POUWTS). POUWTS are decentralized and small-scale technologies that treat water at the home or institutional (e.g., school, hospital) level. They are thus often regarded as valuable for addressing WaSH inequities in the immediate term as no additional infrastructure is required for their distribution. One may further be considered a SMDWS if its effluent meets international drinking water guidelines, as JMP availability and accessibility requirements are otherwise met by its nature (WHO; UNICEF 2017, WHO 2011). However, if a POUWTS is unused or misused after distribution, JMP water quality criteria will not be satisfied. Inclusion within the SMDWS paradigm is therefore predicated on its long-term usage, in addition to its technical water treatment and production capacities.

Selecting a POUWTS that meets these social, environmental, and technical criteria within a given context is therefore critical.

Box: A Brief History of POUWTS

In 1973, E. F. Schumacher penned *Small is Beautiful*, in which he proposed the development of "intermediate technology" (IT) (Schumacher 1973, C. P. Sianipar, et al. 2013). He was responding to a wave of technological critique known as *inappropriate technology theory*, in which scholars argued international development practices relied too heavily on technology that failed to align with the conditions present in the "non-modern sector" of countries "in the early stages of industrial development" (James 1980). The concept of IT was moreover premised on Ragnar Nurkse's supposition that lower-income communities would benefit from "simpler tools and equipment [that] may be appropriate to the relative factor endowments of countries of this type" (Nurkse 1953). Schumacher thus intended to accelerate social and economic development through the implementation of technology whose "sophistication" and "means of production" matched that of where it was being implemented; as he put it, "production from local resources for local needs is the most rational way of economic life" (Schumacher 1973). IT was, however, initially rejected by mainstream economists. That is, despite the clear ontological rooting in contemporaneous modernization and capitalist theories, IT was widely viewed as propagation of Schumacher's socialist perspective (Varma 2003). It was only in the 1980s that Barbara Ward's famed 'basic needs' took hold of international discourse and IT – now renamed AT – became central to development programming (Murphy, McBean and Farahbakhsh 2009, Lorenzini 2019d, Black 1998). POUWTS resultingly increased in popularity thereafter as researchers around the world drew upon Schumacher's ideas to develop small, low-cost, and decentralized solutions for those in need of safe water (C. P. Sianipar, et al. 2013).

3.1 POUWTS User Preferences, and CWF Selection

Studies by Albert et al. (2010) and Luoto et al. (2012) were among the earliest such works to investigate factors leading to POUWTS sustainability from a user perspective. Namely, the researchers distributed POUWTS to 400 households in Kenya and 800 households in Bangladesh, respectively, after which participants used one technology for two months and then switched to use another for the same time period until all had been tested. CWFs were evaluated alongside different chlorinated disinfectants (e.g., Aquatabs, WaterGuard). In Kenya, the largest proportion of participants (45%) stated they preferred the CWF over other options, of whom 90% cited its ease of use as the primary reason. Chlorine products were widely noted as unpreferable due to factors including difficulty of use, perceived ineffectiveness, and residual tastes (Albert, Luoto and Levine 2010). In Bangladesh, CWFs were also the most preferred solution and the residual taste and odor from all chemical

disinfectants was a leading cause of disuse and disinterest. However, participants were asked to specify how much they would pay for each product, and more were willing to pay market price for chlorine solutions. Specifically, CWF price was vocalized as too high given their slow filtrate production; this perspective remains prevalent within more recent literature as well (Burt, et al. 2017). These studies thus demonstrated that technological capacity, simplicity, and effluent aesthetics are important for promoting usage, but cost remains critical to whether it is acceptable in the first place within the existing capitalist model of POUWTS distribution in the global South.

Santos et al. (2015) and Pagsuyoin et al. (2015) later expanded from these insights by quantifying POUWTS sustainability and appropriateness according to "Analytical Hierarchy Process" (AHP) modeling. Six POUWTS (solar disinfection, water boiling, chlorination, disinfection-flocculation with *Moringa Oleifera*, CWFs, and biosand filters) were evaluated in the rural Philippines according to fifteen criteria related to environmental sustainability, technical efficacy, ease and cost of use and maintenance, material availability, and social acceptance. Approximately 100 individuals participated in a multi-day workshop and tried each of the POUWTS options before ranking them in each category according to a defined rubric. Results demonstrated that Moringa was the optimal solution of those evaluated, whose natural abundance in the study area led to a high social acceptability. CWFs were the second most preferable solution, largely due to the ease of use and pre-existing availability within the given community. The cost and slow flowrate were both, however, identified as significant deterrents, particularly when related to the free and already widely used Moringa.

These studies together provide important knowledge related to a POUWTS' appropriateness. That is, CWFs are an easily used and thus preferable solution to individuals living in highly

diverse contexts, suggesting it may hold a degree of universal acceptability that could promote adoption. Further exploration of its field performance is required to better understand its ability to be integrated within the SMDWS paradigm. That said, while ease of use is important in terms of user interest, availability, cost, and especially familiarity are also critical in determining whether a POUWTS is sustainable within a specified context. These perspectives were moreover particularly relevant to the development and analysis of this work's consultation phase described in Chapter 7.

3.2 CWFs and Health

One matter that was notably absent from the abovementioned studies was evaluation of POUWTS usage/adoption and associated health impacts over long time periods (i.e., >1 year). That is, significant scholarship has illustrated that short-term and long-term technological usage, particularly as it relates to health, are phenomena which follow differing psychological processes (Straub 2009, Middleton, Anton and Perri 2013). While important factors related to POUWTS acceptability in the immediate term were thus identified, factors related to long-term acceptance, and how that acceptance impacts health, remain fairly unelucidated. The prevalence of this challenge is further particularly acute within CWF research, as usage has continually been shown to decrease with time. For instance, du Preez et al. (2008) found 45% of interviewees stopped using a CWF after 6 months of ownership while Brown et al. (2009) reported 91% of interviewees stopped using a CWF after 42-48 months. A systematic review further found as many as 93% of participants stopped using a POUWTS within 6 months of implementation (Martin, et al. 2018). As such, as summarized by Wolf et al. (2014), research demonstrating "people's acceptance, adoption, and sustained use [of a POU] is still rare."

Consequently, the related long-term impacts of CWFs on health are unclear. This ambiguity is exemplified within the metanalysis conducted by Clasen et al. (2015), who evaluated several POUWTS including CWFs. Of the 161 studies included, none had an intervention evaluation period that extended beyond 12 months. Additionally, within these studies, associated long-term health outcomes were not well understood. For instance, the authors note that approximately 60% of CWF users observed an associated decrease in diarrheal illness after intervention. However, the usage and health data extracted from the published literature was so variable with time that the analytical response model yielded confidence intervals too large to make justifiable inferences. A positive relationship between CWF usage and health was therefore suggested, but additional research investigating long-term (i.e., multi-year) sustainability and health is needed for such an affirmation to be confirmed.

Peletz et al. (2012) also made similar observations, though at a more granular level. Namely, the authors found 96% of households in Zambia owned a working CWF after 12 months, yet only 75% reported using it. Further, of those users, only 50% reported positive associated health impacts, while more than 60% had total coliform levels in their drinking water that were deemed "high-risk". As such, challenges with filter disuse, efficacy, and diarrhea were observed over time. Comparable results were also shown by Brown (2007) who found no statistically significant relationship between measured effluent *E. coli* levels and reported health improvements over 44 months in Cambodia. And though most households using a filter at the time of the survey reported a positive health impact due to its usage, a measured 2%/month CWF disuse rate limited the outcomes' statistical power. The specifically long-term impacts of filter intervention were thus unconfirmed.

Mellor et al. (2014) later illustrated related findings when using collected data on CWF efficacy, CWF usage/maintenance, and diarrheal episodes in South Africa to develop a predictive model of these same outcomes. Filters removed between 90% and 99.99% of total coliforms at the time of implementation, which changed to -99% to 99.99% by the 3-year time point; in other words, some filters released more microbes than they removed. Further, if filters removed 99.9% of total coliforms or more, 0-2 cases of diarrhea were estimated per person per year. When CWFs removed less than 99% (the WHO guideline for household water filters), 4-8 cases of diarrhea were estimated. Additionally, a second crucial element of CWF impact was how it was used and maintained. 8-10 diarrheal cases were predicted among participants who did not report using the filter daily and 5-7 cases were estimated among those who did. 2-4 diarrheal cases were also estimated among users who reported cleaning their filter more than once every 4 months, whereas 3-6 cases were estimated among those who cleaned their filters once per year or less. As such, long-term CWF impact was highlighted as not only related to filter treatment efficacy but also the way in which it was used. As put by the authors, "broadly defined human behaviours [were] a primary driver" of diarrheal illness. Though, due to the 1- and 2-year intervals between monitoring points, measurements were highly heterogeneous and generalizable inferences were difficult to ascertain. Research into how health and behaviours change with time, wherein more frequent participant engagement and improved usage/maintenance support is provided, is therefore needed to better understand the health-related value of a CWF as a SMDWS.

3.3 Psychology of Behaviour Change

CWF and POUWTS usage behaviours are furthermore demonstrably related to the impact on diarrheal illness, making research into facilitating the development of such behaviours

critical. This need is particularly well captured by Meierhofer et al. (2018) who found simple activities like washing a filter and/or bucket with a household cloth significantly worsened effluent water quality while wearing a different pair of shoes to use the toilet significantly improved effluent water quality. Diverse scholars have thus posited various theories to guide behaviour-targeting interventions, though *persuasion theory* – originally proposed by Sherif and Cantril (1947) – remains among those most popular (Fiebelkorn, et al. 2012, Dreibelbis, et al. 2013, Ginja, Gallagher and Keenan 2021, C. P. Sianipar, et al. 2014, Martin, et al. 2018).

These psychologists argued that attitudinal and behavioural change are functions of the ego and one's self-concept, meaning one's proclivity towards being persuaded to change is based upon how that change aligns or misaligns with one's self-image. The theory therefore suggests that implementers must address matters of identity more than cognition and informational knowledge. Research by Kraemer and Mosler (2010) later illustrated the relevance of this theory to POUWTS when evaluating an intervention in Zimbabwe. Namely, nine factors were considered: involvement (i.e., in decision making), attitude, habit, affect (i.e., emotional perception of a technology), knowledge (i.e., understanding of purpose/methods), beliefs, perceived benefit, ability (i.e., ease of use), social influence, and self-persuasion (i.e., one's adoption of a behaviour becoming a motivator to continue it). Participants were then asked questions related to their experiences and behavioural tendencies after 3 weeks of intervention, and social influence and self-persuasion were noted as most impactful on whether someone was willing to use the POUWTS. A change in one's habit was thus considered more related to how one feels about themselves personally and as a member of their community than the knowledge of whether such a habit is *'good'* or not.

One shortcoming with this work, however, is that long-term habits were not considered. Even further, no research is known to have evaluated these principles over longer time periods in a POUWTS and/or CWF context. With that said, diverse psychological literature suggests one maintains a behaviour if they are satisfied with how their life has changed after doing so. Put another way, behavioural adoption is a response to the difference between experience and expectation, while behaviour change maintenance is the response to the difference between expectation and fulfillment (Chaiken 1980, Rothman 2000, Cismaru, Nagpal and Krishnamurthy 2009). A significant gap in the literature therefore exists regarding how persuasion theory is applied to behaviour change maintenance. And more acutely, additional research is needed regarding how to facilitate that maintained change in water-related habits.

Specifically, while scholars commonly suggest that knowledge communication is crucial to promoting certain behaviours (Dreibelbis, et al. 2013, Ginja, Gallagher and Keenan 2021), limited research exists which exemplifies how to do so, and what impact it has. For instance, Tamas and Mosler (2011) promoted SODIS for two months to groups in Bolivia but found 70% of participants stopped using it after intervention. Alternatively, after Meierhofer et al. (2018) observed behavioural challenges, they provided one-to-one education to participants but found "the change in awareness achieved by the training did not sufficiently result in adequate change in their practices." Ginja et al. (2021) responsively notes how these uncertainties are due to "existing approaches [having] overemphasized hypothetical psychological variables," later recommending that "interventions should, amongst others, provide necessary how-to-do knowledge" as well as "alter cues in the environment so that habits can be formed through behaviour repetition and reinforcement." Meanwhile, specific methods related to how to do so are understudied. Beyond education itself, exploration how

and how much information should be relayed, as well as what information should be included to facilitate long-term social acceptance, is required.

3.4 Participatory and Decolonizing Methods

Among the most highly regarded and widely referenced frameworks for including the social within WaSH technology interventions are decolonizing and participatory methods. Decolonizing methods gained particular popularity during Indigenous rights movements in the late 1990s and early 2000s (D. Hodgson 2011a, G. Adams 2014). These and other marginalized populations had lamented the historically extractive and exclusionary processes involved in program development and associated knowledge creation. Advocates thus detailed a need for reform that centred the voices of intended benefactors within project inception, production, implementation, and assessment. Meanwhile, participatory methods emerged concurrently and drew from similar principles, centering local knowledge and ontology within intervention programming (Hazeltine 2003, Harmacioglu 2017, WHO 1996). It is noteworthy that various academics and advocates have regarded participatory scholarship as a *Westernization* of decolonizing methods, intended to maintain its insights while erasing the specific relevance of colonialism and neocolonialism to existing power inequities (Pohlhaus 2012, Richardson 2019, Chouinard 2016). Nevertheless, participatory methods remain among those most commonly referenced within the WaSH sector (WHO 1996, WHO 2020, UN 2015).

Described briefly, participatory and decolonial methods are approaches founded upon principles of promoting dialogue, including all project stakeholders within the various elements of the program cycle, seeking to build on existing community strengths and knowledge, iterating processes cyclically to respond and adapt, and committing resources

over the long term (Blumenthal, Hopkins III and Yancey 2013). As put by Kindon et al. (2007), the strategy "treats participants as competent and reflexive agents capable of participating in all aspects of the research process" to "integrate values and beliefs that are indigenous to the community into the central core of interventions and outcome variables." Using the language of decolonial scholarship, the methods invoke "consciousness of neocolonial oppression as an enduring force in the modern global order," to "stand with people in 'other' settings to come to a better understanding of relationality" (G. Adams 2014). The intention is moreover to decenter implementing bodies and recenter participants such that program outcomes are determined by community members for community members in need of assistance, without pollution by external biases.

Yet, despite its significant prevalence in WaSH discourse (most notably SDG 6.a), a relative dearth of data exists which demonstrates specific participatory implementation methods, or how they may translate to associated improvements in programmatic efficacy (Wolf, et al. 2014, Ginja, Gallagher and Keenan 2021, Ray and Smith 2021). For instance, the most widely used framework in WaSH is the WHO's Participatory Hygiene and Sanitation Transformation (PHAST), which is founded on a behaviour change strategy called Self-esteem, Associative Strengths, Resourcefulness, Action-planning, and Responsibility (SARAR) (WHO 1996). A study in Senegal that incorporated this program as the kernel of its implementation strategy, however, found that after 4 years, the program's "behaviour change appeared to be insufficient to establish […] habits and norms" (USAID 2019). As one example, only 27% of participants continued to use one of several POUWTS, of whom the majority used one which was not included in the intervention strategy. As such, while participatory principles are prevalent and valuable, the WaSH sector remains challenged in

converting these principles to actionable methods that produce improved results. Additional research into participatory intervention planning and implementation is needed.

3.5 Summary of Research Needs

CWFs offer significant opportunity to advance access to SMDWS in the immediate term, yet researchers and practitioners have been challenged in promoting their long-term usage and achieving associated positive health impacts. Evidence from literature has shown familiarity and social acceptance are important determinants of POUWTS/CWF adoption. Participatory research principles have further gained popularity within the WaSH sector as valuable in facilitating habit formation and promoting this associated technological acceptance. However, practical methods that can promote participation such that CWF adoption and water-related health outcomes are improved remain unclear. For instance, limited research has evaluated long-term (>12 months) CWF usage and resulting diarrheal health, and of those which have, none have engaged with users frequently enough to gain insight into how behaviours and health change with time. Participatory implementation, wherein participants are actively engaged in project progression and knowledge creation processes, is thus understudied. Similarly, WaSH scholars commonly reference knowledge communication as a key element of participatory programming and an important feature of any technological intervention. But the frequency, duration, and content of such communication, as well as means of ensuring it is participatory, locally relevant, and persuasive, is largely unexplored. Work is therefore also required to evaluate methods of long-term knowledge communication and the associated CWF impacts within a participatory framework. Identification of key factors influencing the relationship between knowledge communication, technology usage, and water-related health is needed.

Chapter 4: Silver and Zinc Oxide Nanoparticle Disinfection in Water Treatment Applications: Synergy and Water Quality Influences

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Abstract:

The synergistic potential of silver and zinc oxide nanoparticles for water disinfection was investigated herein. By causing cell death through membrane interactions, oxidative killing, and DNA deactivation, metallic nanoparticles may be integrated with point-of-use water treatment systems for applications in rural and remote geographies. Disinfection efficacy was evaluated in batch-phase experiments under both synthetic and real water conditions, where synthetic water was varied by pH and dissolved oxygen levels. Ceramic pot filters with comparative nanoparticle concentrations were also investigated. In all cases, combinations of silver and zinc nanoparticles resulted in improved disinfection in comparison to either metal in isolation. In batch experiments, dissolved oxygen proved to be particularly impactful, with kinetic rates reducing approximately 45% when in low oxygen environment (< 3 mg/L) versus high oxygen (>8 mg/L). Log removal values (LRV) were further, on average, 31% lower in real water than synthetic water after 300 minutes, though silver-zinc combinations were still superior to either metal alone. In filters, those impregnated with 67% silver, 33% zinc achieved average LRV's of 2.7 and 2.9 after 60

minutes of filtration and 24 hours of storage, respectively, while those with only silver achieved average LRV's of 2.0 and 3.1 at those same times.

Keywords:

Ceramic Water Filters, Drinking Water, Low-Cost Disinfection, Metallic Nanoparticles, Rural Water Supply

Highlights:

- Combining silver-zinc oxide nanoparticles improves disinfection in batch-phase and ceramic pot filter experiments.
- Nanoparticle disinfection is vulnerable to changes in water quality, and particularly dissolved oxygen.
- Zinc oxide supplementation of silver simultaneously improves effectiveness and reduces costs of water treatment solutions.

4.1 Introduction

Nearly 1500 children under five years young died from diarrheal diseases every day of 2017, due in large part to drinking water that is microbially contaminated (WHO & UNICEF 2019, WHO 2017). These deaths, however, were not spread evenly across nations, but were concentrated in those with the world's poorest and most vulnerable populations. Specifically, 96% occurred in UN-defined *Developing* nations, compared with 0.03% in those defined as *Developed*, illustrating the clear relationship between health and economic status (UNICEF 2015). Further, infant mortality, and diarrhea particularly, impacts rural communities in much greater numbers than their urban counterparts (Poel, O'Donnell and Doorslaer 2009, Yaya, et al. 2019), exacerbating the social and economic inequities that maintain a system of marginalization (UNECOSOC 2017, World Bank 2016).

Point of use water treatment systems (POUWTS) have moreover been highlighted as a solution to address concerns with safe water access in rural contexts, with a diversity of options available on the international market (Santos, Pagsuyoin and Latayan 2016). And while chlorine remains the most popular choice in developed systems for disinfection, access in rural areas is often more challenging. Further, there are persistent complaints from individuals who find POU chlorination products unappealing due to the residual taste they leave in water, which then leads to disuse; in addition, the necessitated recurring cost of single-use chlorine products (approximately $0.10 USD/week) have also proven problematic (Sobsey, et al. 2008, Luoto, et al. 2012, Santos, Pagsuyoin and Latayan 2016).

One set of alternatives that has garnered notable attention for enhanced disinfection is metallic nanoparticles, due in large part to the weight to volume ratios achieved at such a small scale (Qu, Alvarez and Li 2013). As such, research into nanoparticle disinfection has included applications in conventional centralized drinking water and wastewater treatment processes, as well as home-based, point-of-use water treatment technologies (Prathna, Sharma and Kennedy 2018, Qu, Alvarez and Li 2013, Slavin, et al. 2017). Silver nanoparticles (AgNPs) are the most commonly investigated metal species due to their long history as an applicant for medical treatment, as well as their observed superiority to other metals in terms of bacterial disinfection (Malachova, et al. 2011, Al-Issai, et al. 2019, Rai, Yadav and Gade 2009). For example, Al-Issai et al. (2019) investigated silver (Ag), zinc oxide (ZnO), magnesium oxide (MgO), and copper (Cu) for disinfection in reverse osmosis and multistage filtration treated water and showed AgNPs achieved a log removal value (LRV) of 2.43±0.03, whereas ZnO, MgO, and Cu achieved LRVs of 1.80±0.25, 0.77±0.04, and 0.37±0.08, respectively (Al-Issai, et al. 2019).

Concerns, however, remain regarding silver's health, economic, and technical impacts, hindering its applicability to POUWTS. Specifically, over-consumption of silver can lead to negative health outcomes such as Argyria or even DNA damage (Fewtrell, Majuru and Hunter 2017), leading the World Health Organization (WHO) to recommend a 0.1 mg/L limit on silver concentration in drinking water; this concentration ensures less than 10 g of silver be consumed over a 70-year lifespan (WHO 2011). Silver is also a notably expensive material, with a cost of approximately $3 USD per gram (Argenol Laboratories 2017). Though variability exists depending on nanoparticle formulation (e.g., solid or aqueous), as

well as by providers and location, its inclusion in water treatment technologies intended for individual use can nonetheless contribute to such products being unaffordable for low-income consumers in particular, further limiting their widespread applicability as an antimicrobial solution to those most in need (Burt, et al. 2017). Investigations of alternative metal species that can contribute to disinfection efficacy with decreased long-term potential health impacts are thus required.

ZnO has demonstrated promise as a potential alternative due to its own antimicrobial properties under diverse conditions (Dimapilis, et al. 2018, Sirelkhatim, et al. 2015). Further, research has shown that zinc can substantially reduce the incidence and severity of diarrheal episodes when ingested, having a doubly positive effect as a water purification solution (Gitanjali and Weerasuriya 2011, Malik, et al. 2013). Finally, zinc oxide not only has a cost of approximately $0.09 USD per gram, more than 30 times less than that of silver (Sigma-Aldrich 2021), but may be commonly found locally and is thus often more accessible than silver; zinc accessibility should, however, be evaluated on a case-by-case basis.

While silver has been shown to exhibit greater disinfection efficacy than zinc alone (Al-Issai, et al. 2019, Malachova, et al. 2011), investigation into the potential replacement or supplementation of silver with co-nanoparticle application is understudied. Of the limited research available to-date in this area, most findings are intended for medical applications, whereas water treatment applications, and particularly applications within low-cost water treatment technologies, remain largely unelucidated (Garza-Cervantes, et al. 2017, Motshekga, et al. 2015). For example, Garza-Cervantes et al. (2017) found 32.7 mg/L of Zn^{2+} combined with 3.21 mg/L Ag^{+} achieved 100% *E. coli* growth inhibition after one hour,

compared with approximately 30% *E. coli* growth inhibition with Ag^+ alone. The focus of this work on bacteriostasis within culture media for medical purposes does, however, lend itself to questions regarding how co-metal disinfection may be applied in an aquatic environment for bactericidal water treatment. Similarly, Motshekga et al. (2015) examined bacteria removal based on a combined solid phase adsorption and disinfection and reported a silver-zinc disinfection synergy, but their work did not examine direct nanoparticle interventions under drinking water applications, or if such findings may be translated to technology. Finally, some recent research has also highlighted the potential of co-metal disinfection within technological applications, however the limited literature in this domain has focused particularly on silver-copper and silver-iron combinations with variable results (Guerrero-Latorre, et al. 2015, Brown and Sobsey 2009, Ehdaie, Su, et al. 2020, Lucier, Dickson-Anderson and Schuster-Wallace 2017). An important research gap thus remains in terms of how combined metal nanoparticles, and silver-zinc combinations specifically, may synergistically disinfect bacteria within a water treatment application, and particularly within point-of-use water treatment technology.

In this research, Ag and ZnO nanoparticle disinfection of *E. coli* is investigated in isolation and in combination to evaluate their respective and joint antibacterial efficacies for water treatment applications. Nanoparticles are characterized, and bacterial destruction is observed by Scanning Electron Microscopy. Antibacterial performance in batch-phase experiments is evaluated against several water quality parameters, which is lacking in the literature. Specifically, disinfection efficacy and kinetics with co-metal addition of silver and zinc nanoparticles under varying pH and DO conditions are studied. Further, full-scale ceramic

pot filters with corresponding silver-zinc nanoparticle ratios are investigated for bacteria removal using a real water source to determine technological applicability. Moreover, the research into synergistic silver-zinc metal nanoparticle disinfection for water treatment applications highlights a novel methodology that has the capacity to be easily incorporated into point-of-use technologies, improving water treatment options for lower-income and remote consumers in terms of both effectiveness and affordability.

4.2 Materials and Methods

4.2.1 Scanning Electron Microscopy

Scanning Electron Microscopy (SEM; Nano Imaging Facility, Carleton University, ON, Canada) was used to determine the size range for silver and zinc nanoparticles in both solid and aqueous phases. 1 mg/mL suspensions of each species were created and mixed gently for 5 minutes, and subsequently allowed to sit in an aqueous phase for 5 hours. 1 mL of solutions were then pipetted atop a 0.45 µm filter paper and left to dry inside a desiccator overnight. A small piece of the paper was cut and adhered to a sample mount with double-sided adhesive tape and sputter-coated with gold before entrance into the SEM imaging hood. Energy Dispersive Spectroscopy (EDS) was used to determine which parts of the sample contained the specific nanoparticle material, as well as when both elements were present if the sample was a mixture.

Bacterial imaging under 4 specific challenge conditions was also conducted using SEM. Namely, no metals, 1 mg/L Ag, 1 mg/L ZnO, and 0.67 mg/L Ag & 0.33 mg/L ZnO together were added to 10^5 CFU/mL solutions, which were agitated at 50 RPM overnight.

Dehydration and fixation then followed a modified procedure described by (Iqbal, Lai and Avis 2012). Briefly, after agitation, samples were gently mixed by hand and poured over 25 mm diameter 0.2 µm nylon filter papers (Millipore). The filter papers were then placed within marked aluminum dishes and suspended within a 4% glutaraldehyde mixture within a 0.1 M phosphate buffer solution (75% Na_2HPO_4, 25% NaH_2PO_4) for 60 minutes. After 60 minutes, the glutaraldehyde solution was filtered again atop the papers, which were then twice resuspended in a 0.1M PBS wash for 10 minutes. After washing, a 1% tannic acid solution within a 0.1 M PBS buffer was added and papers were suspended within the solution for 1 hour. After said time, the tannic acid solution was filtered through the papers, which were then washed twice with distilled water for 10 minutes. The dehydration process commenced thereafter: papers were sequentially suspended within ethanol (EtOH) solutions of the following concentrations: (1) 35% EtOH, (2) 50% EtOH, (3) 75% EtOH, (4) 95% EtOH (x2), (5) 100% EtOH (x2). After dehydration, the aluminum dishes containing the papers were placed in a dessicator and allowed to dry overnight. A small piece of the filter papers was then cut with a specialized hole puncher and mounted on an SEM stub with double-sided tape. The samples were then sputter coated with gold and loaded into the SEM viewing cabinet.

4.2.2 Nanoparticle Characterization

Before suspension in water, silver nanoparticles have sharp edges and rigid bodies, ranging from approximately 100 to 600 nm in size. As shown in Figure 4.1A, after suspension for 5 hours, AgNPs dissolved into smaller particles ranging from 34 to 58 nm or agglomerated to form larger particles up to approximately 2 µm in diameter. ZnO nanoparticles are box-

shaped rods before suspension in water, with diameters ranging from approximately 40 to 440 nm. After suspension, the shape of the particles do not change significantly, though agglomeration and dissolution occurs and particle sizes change slightly to be between 30 and 680 nm (see Figure 4.1B). These results align with previous observations from literature that noted ZnO is relatively stable in water and thus does not rapidly dissolve (Sirelkhatim, et al. 2015).

When silver and zinc are added to water simultaneously, ZnO appears to agglomerate around the Ag-NPs and form large mixed particles (Figure 4.1C), as confirmed by EDS analysis (data not shown). Specifically, silver was only detected in the presence of zinc, and in a concentration of approximately 5% of the spectra, compared with approximately 70% for zinc.

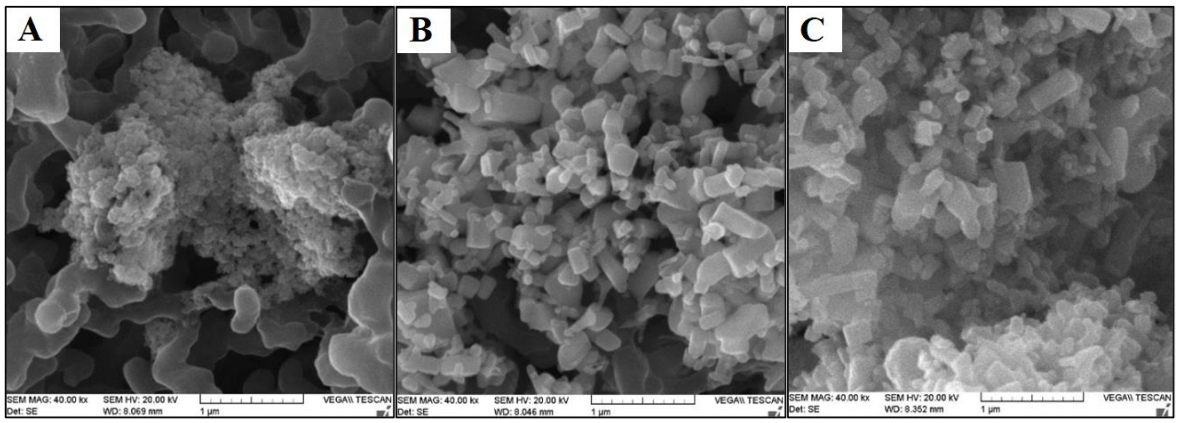


Figure 4.1. Scanning Electron Microscopy images of (A) silver nanoparticles after suspension in water, (B) zinc oxide nanoparticles after suspension in water, and (C) both zinc oxide and silver nanoparticles suspended in water in even molar proportions.

4.2.3 Challenge Water Solutions

Both synthetic and natural waters were examined in this research. Synthetic challenge water was created as per Table 7 of USEPA "Methods for Measuring the Acute Toxicity of

Effluents and Receiving Waters to Freshwater and Marine Organisms" (USEPA 2002), intended to match the alkalinity of the batch-tested natural water source. Natural water for batch testing was taken from Ottawa River at the Britannia Water Treatment Plant intake (Ottawa, Canada), while ceramic pots were challenged by natural water drawn from a nearby stream to the Nelson Mandela African Institute of Science and Technology (NM-AIST) in Checkereni, Arusha, Tanzania. Note that the batch-scale evaluation of natural water outcomes are demonstrative of matrix complexity impacts, particularly with respect to dissolved organic carbon (DOC). A summary of the synthetic water and the natural water quality conditions are presented in Table 4.1.

pH was adjusted before testing to approximately 6.4, 7.4 and 8.4 using 1N HCl and NaOH solutions as needed, described as Low pH, Mid pH, and High pH in Table 4.1, respectively. Low DO testing water was deoxygenated with compressed N_2 gas sparged into the water to maintain oxygen levels just above hypoxic conditions. These specific water quality parameters were chosen to align with the theoretical basis by which metallic ions and ROS are released by nanoparticles, as per either of Reactions 4.1 or 4.2.

$$NP_{(s)} + O_{2(\mathrm{aq})} \rightarrow \mathrm{ion} + {}^{\bullet}O_2^- \qquad [4.1]$$

$$NP_{(s)} + \frac{1}{2}O_{2(aq)} + 2H^+_{(aq)} \rightarrow 2ion + H_2O_{(l)} \qquad [4.2]$$

where *NP* represents either AgNP or ZnO and *ion* is the ionic version of the associated metal in reacting (either Ag^+ or Zn^{2+}).

Table 4.1. Water quality characteristics of experimental conditions evaluated.

Water Quality Characteristics	Synthetic Water						Natural Water	
	Low pH, Low DO	Low pH, High DO	Mid pH, Low DO	Mid pH, High DO	High pH, Low DO	High pH, High DO	Ottawa River Water (Canada)	Checkereni Stream Water (Tanzania)
pH	6.40±0.03	6.41±0.03	7.39±0.02	7.38±0.06	8.39±0.02	8.41±0.01	7.32 ±0.10	8.07±0.11
DO (mg/L)	2.51±0.40	8.04±0.25	2.50±0.06	8.20±0.28	2.51±0.04	8.45±0.06	10.34±0.89	7.64±0.59
ORP (mV)	13±2	31± 2	-48±2	-41±2	-109±2	-96±7	-41±5	-55±6
Alkalinity (mg/L $CaCO_3$)	23.7±6.7	30.2±5.8	31.2±2.8	32.7±4.2	30.2±2.5	37.85±5.90	38±2	NA
Temperature (^{O}C)	20.5±0.5	22.6±0.6	20.9±1.0	21.9±0.6	22.3±0.7	21.7±0.2	20.8±1	21.3±1.2
Initial E. coli Concentration (10^5 CFU/mL)	8.61±1.11	3.26±1.16	7.44±1.72	3.98±2.28	9.30±4.94	5.84±2.76	8.89±2.76	0.25±0.42

Note: DO = Dissolved Oxygen; ORP = Oxidation-Reduction Potential; CFU = Colony Forming Units

4.2.4 Bacterial Cultivation and Enumeration

K-12 (ATCC® 29947) *Escherichia coli* (Migula) Castellani and Chalmers was purchased from Cedarlane laboratories as a dry pellet. The pellet was hydrated with 1 mL of No.3 Nutrient Broth and centrifuged for 10 seconds. 200 µL of *E. coli* stock was added to 9800 µL of nutrient broth and incubated at 37°C for one hour. 1 mL of sterile, pure glycerol was added to the stock solution, after which the stock was incubated again for 3 hours and subsequently divided into 200 µL aliquots, which were stored in 2 mL sterile tubes at -20°C. For testing purposes in batch experiments, the frozen stock was thawed at room temperature for 5-10 minutes and 100 µL was added to 9900 µL of No.3 Nutrient Broth and incubated at 37°C for 18-19 hours, the beginning of the stationary growth phase. 1 mL of this final culture was then added to 1 L of challenge water, yielding approximately 10^5 CFU/mL initial bacterial concentration. It is noteworthy that K-12 *E. coli* is known to have a thinner cell membrane than others found in the natural environment and may thus be more susceptible to disinfection impacts. The present methodology must therefore be considered a demonstration of concept rather than an illustration of what is to be expected within a field setting. Additional research evaluating diverse bacterial strains and species is further recommended.

For pot experiments, faecal droppings from goats shepherded near NM-AIST were collected and submerged in water from the stream for 1-2 hours before testing to achieve elevated levels of *E. coli* contamination than was present immediately after extraction.

E. coli concentrations during testing were enumerated as per USEPA Method 1604 (USEPA 2002). 100 µL was taken from the experimental vessel at each measurement point and added to 900 µL of Dilution Water (0.5% $MgCl_2$, 0.125% KH_2PO_4) in a sterile, 2 mL conical vial. The sample was then diluted serially up to 4 steps and samples were filtered through a 47 mm diameter, 0.45 µm pore membrane filter paper. The paper was then placed atop MI Agar Media (Thomas Scientific) and incubated for 24 hours. Colonies were counted under black light by hand.

4.2.5 Disinfection Performance of Metal Nanoparticle Species

Disinfection efficacy and kinetics of silver and zinc nanoparticles in batch experiments were evaluated in both a synthetic and natural water. The pH and DO levels were varied within the synthetic water matrix as illustrated in Table 4.1. The natural water was not adjusted. Prior to testing, 1 mg/mL solutions of AgNPs (Argenol Laboratories, Spain) and ZnO (Puriss, >99% (KT), Sigma-Aldrich) were created, which were subsequently added to 1 L of challenge water in the appropriate volumes to yield concentrations ranging from 0 – 1 mg/L (see to Table 4.2). *E. coli* samples were collected at 7 time points: 0, 10, 20, 30, 60, 120, and 300 minutes after metals were added to the challenge water. All samples were taken in duplicate. All water chemistry conditions and metal combinations were also tested twice (i.e., n=4 per sample). Challenge water was constantly mixed at 50 RPM with two sterilized flat-

blade propellers. Before experimentation, beakers were sterilized in an autoclave and wrapped on the outside with aluminum foil to prevent light penetration and ensure dark conditions throughout testing.

Table 4.2 Metal nanoparticle concentrations and naming convention

Metals Added to Challenge Water		Naming
Massive Concentrations	*Molar Concentrations*	**Code**
No metal control	No metal control	NM
1 mg/L ZnO	12.3 µM ZnO	ZnO1
1 mg/L Ag	6.5 µM Ag	Ag1
0.67 mg/L Ag, 0.33 mg/L ZnO	4.4 µM Ag, 4.1 µM ZnO	Ag67ZnO33
0.33 mg/L Ag, 0.67 mg/L ZnO	2.2 µM Ag, 8.2 µM ZnO	Ag33ZnO67
0.14 mg/L Ag, 0.86 mg/L ZnO	0.9 µM Ag, 10.1 µM ZnO	Ag14ZnO86

As demonstrated herein, a log-linear relationship between disinfection and time was observed for all metal nanoparticles evaluated. As such, the Chick-Watson Model for disinfection shown in Equation 4.2 could be applied.

$$\ln\left(\frac{N_t}{N_0}\right) = -kCt \qquad [4.2]$$

Where N_t is the *E. coli* concentration (CFU/mL) at time t, N_0 is the initial *E. coli* concentration, k is the reaction rate constant (hrs^{-1}), C is the concentration of disinfectant (mg/L), and t is time in hours. Further, because all of these experiments were conducted with 1 mg/L of disinfectant, Equation 4.2 may be rewritten as Equation 4.3, allowing for the enumeration of a kinetic rate. Both LRVs and kinetic rates are thus used for evaluation.

$$k = -\left(\frac{1}{t}\right) 2.303 \log\left(\frac{N_t}{N_o}\right) \qquad [4.3]$$

4.2.6 Ceramic Pot Filters

4.2.6.1 Fabrication

Pot filters were fabricated at the Wine to Water East African (W2WEA) facility in Arusha, Tanzania. Clay and sawdust (pine wood) were first mixed in 5:4 (C:SD) volumetric ratio until homogeneous. Silver, zinc, or a mixture of the two was then impregnated within the filters via the co-firing method, where nanoparticles were dissolved into water and added to the dry ingredients to achieve a total metal concentration of 7.5 mg/kg filter. The wet mixture was then kneaded, shaped using a manual press, and labeled. Filters were thereafter left to air dry for 3 weeks, after which they were fired in a kiln up to 960 ^{o}C over 12 hours. Each filter then passed 20 L of clean water through the matrix before bacterial testing.

4.2.6.2 Experimentation

In addition to no-metal controls, filters were produced with metal mass ratios of: (1) 100% Ag, (2) 67% Ag, 33% ZnO, (3) 33% Ag, 67% ZnO, and (4) 100% ZnO. Three filters were produced in each metal ratio, and each filter was tested twice (i.e., n=6). Any filter with a measured flowrate greater than 4 L/hr was, however, excluded, as per the exclusion criteria for sale used by W2WEA. During testing, filters were filled to achieve a hydraulic head of 25 cm above the frustrum (pot bottom), which was maintained for 60 minutes. Filtered samples were collected from the storage container after 30 and 60 minutes of filtration, as well as after 24 hours of storage in a darkened space. Influent water quality was evaluated before challenging the filters, whereas the filtered water quality was evaluated only after 60 minutes of filtration and after storage. Figure 4.2 illustrates a schematic of the experimental setup.

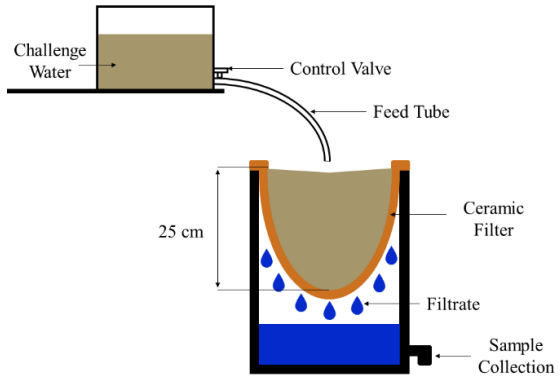


Figure 4.2 Schematic of Ceramic Water Filter Experimental Setup

4.2.7 Statistical Analysis

4.2.7.1 Nanoparticle Disinfection

The significance of each of the treatments described was evaluated through multiple two-way ANOVAs. For experiments conducted in synthetic water, differences in LRV and kinetic rates achieved by the metal nanoparticle combinations were compared across differing pH and DO conditions at each individual measurement time. The impact of natural water on these same response variables was evaluated similarly, where nanoparticle performance was compared against that in synthetic water of the same approximate pH level (pH = 7.4). Post-hoc analysis using Tukey's Honest Significant Difference (HSD) was used thereafter to determine specific differences between treatments. All statistical analysis was conducted in Rstudio (stats and base packages). Tukey's HSD tables may also be found in Appendix E.

4.2.7.2 Ceramic Pot Performance

Differences between filters were also evaluated using two-way ANOVAs, where time and metal concentrations were used as treatment variables and LRV and percent bacteria removal were each used as measured responses. This was chosen to account for the influence of variability in the initial bacterial concentration of the natural challenge water, as highlighted in Table 4.1. Post-hoc analysis using Tukey's HSD as well as 1-tailed t-testing, was used for evaluation of individual effects for the pots. Tukey's HSD tables may also be found in Appendix E.

4.3 Results and Discussion

4.3.1 Disinfection in Synthetic Water

4.3.1.1 Comparison of Silver and Zinc Oxide Individually

Initial studies examined silver and zinc nanoparticle disinfection efficacy when used individually under varying pH and DO conditions, and as can be observed in Figure 4.3, silver performance is significantly superior to zinc when water quality conditions are equivalent. For example, at the Low pH, High DO level, silver achieves a disinfection kinetic rate of -0.110 hr^{-1}, while zinc achieves a rate of -0.046 hr^{-1}. Meanwhile, at the High pH level, both disinfection kinetic rates decrease slightly: in High DO water, silver and zinc achieved rates of -0.069 hr^{-1} and -0.036 hr^{-1} respectively, and in Low DO water, they achieved -0.057 hr^{-1} and -0.027 hr^{-1}, respectively. Moreover, when evaluated individually, it is clear that silver outperforms zinc and would be the preferred disinfectant to improve removal of *E. coli* for water treatment.

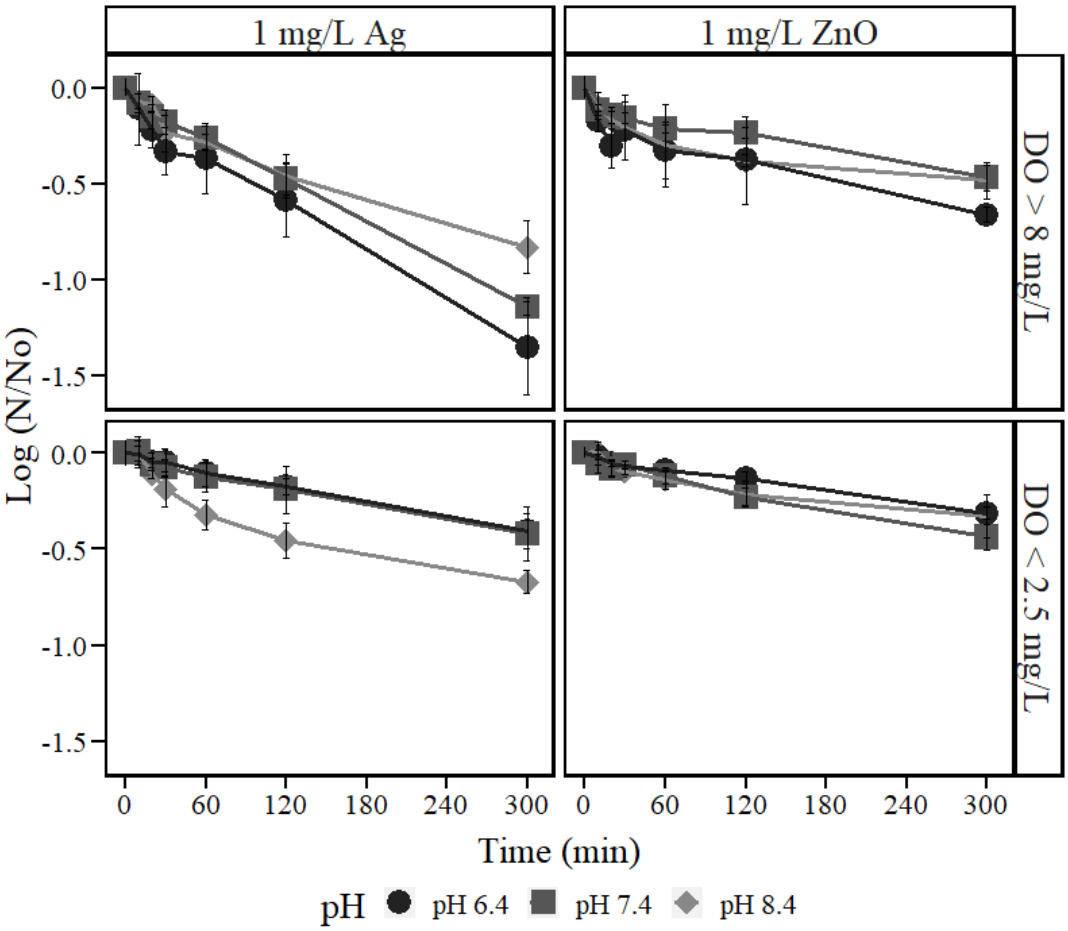


Figure 4.3 LRV for AgNP and ZnO over 300 minutes

4.3.1.2 Comparison of Combined Silver-Zinc Disinfection

Investigations into the combinatorial effect of silver and zinc nanoparticle application in challenge waters varied by pH and DO highlight both a marked difference with the disinfection of each metal when applied individually, as well as a nanoparticle sensitivity to key water quality parameters more generally. Specifically, as shown in Figure 4.4, two clear trends are immediately observed. First, under the High DO conditions, silver-zinc combinations outperform either species in isolation. Further, there is a clear trend where

Ag67ZnO33 continually results in the best outcomes, with LRVs of 3.18 ± 0.23, 3.00 ± 0.22, and 2.91 ± 0.09 after 300 minutes of mixing at pH levels of 6.4, 7.4 and 8.4, respectively.

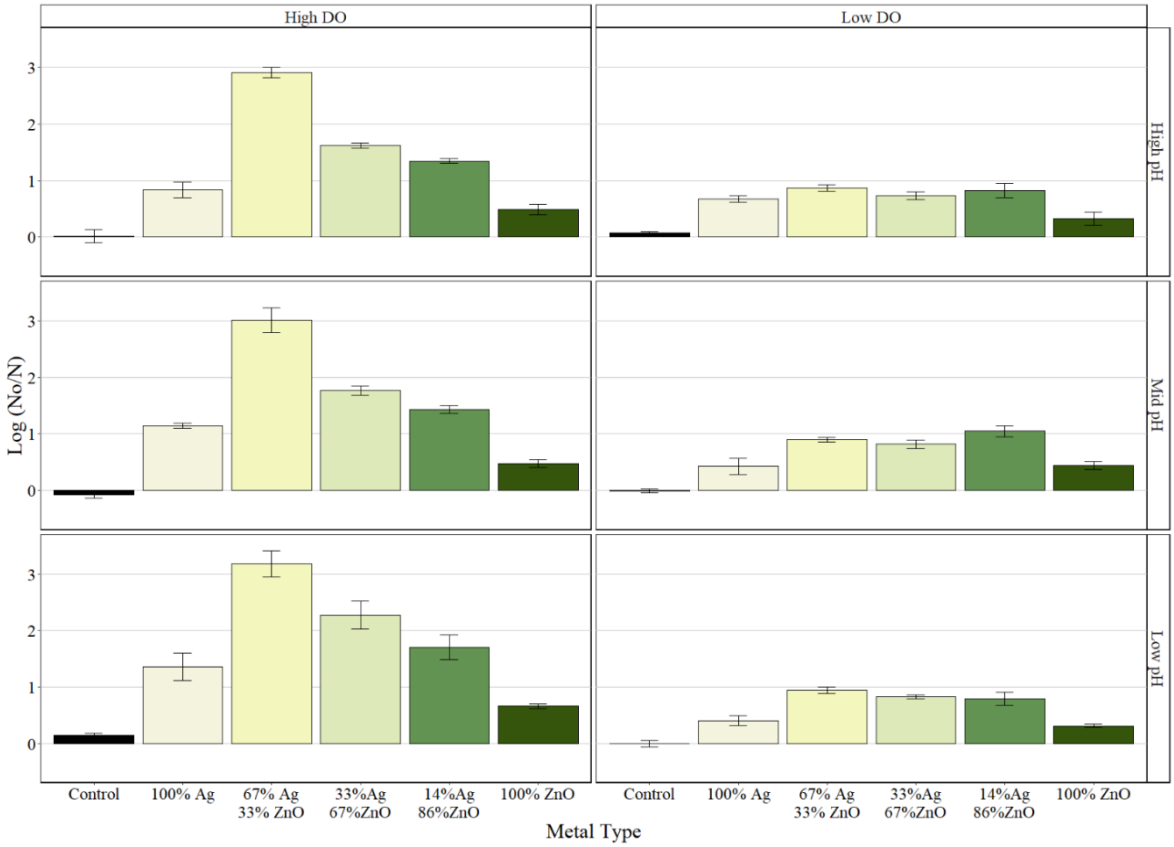


Figure 4.4 Log Removal Values achieved after 300 minutes of mixing under each of the synthetic water quality conditions evaluated and detailed in Table 4.1; pH values of 6.4 (Low pH), 7.4 (Mid pH), and 8.6 (High pH) with High (>8.4 mg/L) and Low (<2.5 mg/L) DO

Secondly, all metallic nanoparticles exhibited a clear and immediate suppression of LRV when High DO conditions are compared with Low DO conditions. For example, as shown in Table 4.3, Ag33ZnO67 achieved a kinetic rate of -0.193 hr^{-1} in the High DO, Low pH condition, dropping to -0.072 hr^{-1} in the Low DO, Low pH condition. With that said, silver-zinc combinations still outperformed either species in isolation (α=0.05), albeit to a reduced degree than observed in the High DO condition. Specifically, as illustrated in Figure 4.5 and

Table 4.3, Ag67ZnO33 achieved a kinetic rate 670.0% higher than zinc alone and 173.3% higher than silver alone in High DO, Mid pH challenge water, but only 102.8% and 101.8% higher than the two species respectively when DO was decreased. Dissolved oxygen was therefore demonstrated to have a profound influence on the disinfection performance of all metals tested, which has particularly important implications for field-based applications of nanoparticles for disinfection. As algal blooms, high faecal matter concentrations, agriculture runoff, and other environmental factors can lead source waters to hover around or fall into hypoxia, the value of the metals may be drastically reduced if exposed to such conditions.

Overall, pH did not significantly impact LRV, indicating some robustness to changing pH levels in a local water source. Others have made a similar observation when examining silver only (Fauss, et al. 2014), though mechanistic investigation of DO and pH impacts on silver-zinc combined performance remains largely unelucidated.

Table 4.3. Kinetic rates achieved by each metal species and combination under each of the water quality conditions evaluated (Bold values indicates highest rate in each experimental condition)

Metal Concentration (1 mg/L Total)	*k (1/hr)*					
Mass Ratio (%) (Ag:ZnO)	Low pH, Low DO	Low pH, High DO	Mid pH, Low DO	Mid pH, High DO	High pH, Low DO	High pH, High DO
100:0	-0.035	-0.110	-0.036	-0.095	-0.057	-0.069
67:33	**-0.081**	**-0.277**	-0.073	**-0.260**	**-0.075**	**-0.250**
33:67	-0.072	-0.193	-0.072	-0.155	-0.063	-0.140
14:86	-0.070	-0.152	**-0.091**	-0.126	-0.072	-0.117
0:100	-0.025	-0.046	-0.036	-0.034	-0.027	-0.036

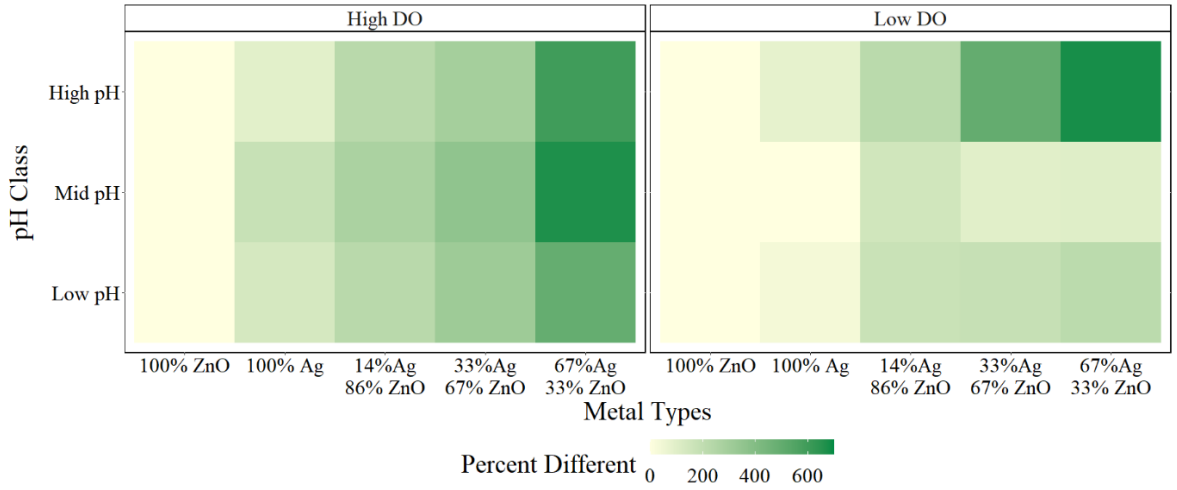


Figure 4.5 Percent difference of kinetic rates from those of 100% ZnO under different pH and DO conditions.

Garza-Cervantes et al. (2017) hypothesize that the mechanism behind the observed synergistic disinfection results from an increased cell membrane permeability observed when nanoparticles are added in combination, in comparison to when added individually (Garza-Cervantes, et al. 2017). Yamamoto & Ishihama (2005) further reported when cells were suddenly "shocked" by an influx of zinc in concentrations higher than 0.065 mg/L (1 μM), zinc-binding proteins began rapid gene transcription to maintain intracellular zinc homeostasis, which subsequently resulted in synthesis of intracellular cysteine, a highly reactive thiolated amino acid. Cysteine production, and the complex and numerous reduction-oxidation (redox) reactions that resultingly ensue, are further known to both increase cell membrane permeability through increasing the active sites available for cationic metal binding (Giles, et al. 2003), and lead to cell degradation directly through transient trapping of metals (Yamamoto and Ishihama 2005). Additionally, McDevitt et al. (2011) found that in *Streptococcus pneumonae*, extracellular zinc in concentrations higher than 1 μM inhibited cells from absorbing other necessary nutrients, weakening the cells and

increasing vulnerability to oxidative killing or bactericide by other present disinfectants, if applicable (McDevitt, et al. 2011).

As zinc concentrations in this research were above 4 μM under all experimental conditions, it is hypothesized that *E. coli* cells were sufficiently 'shocked' by the zinc, leading to transient trapping of zinc within the cell, an increase in active sites along the cell membrane for bonding with thiolated groups (i.e., active sites for cationic metal binding), and a reduction in nutrient intake that weakened the cell. Comparatively, when zinc is in isolation, these effects thus weaken the cells, but the rate of cell degradation may not be as high as when facilitated with the presence of other metals, as was found in this research. When metal combinations were added, cells became more vulnerable to attack from the silver present within the mixture, which may have (1) caused cell lysis directly by bonding with the cell membrane and causing pits, (2) penetrated the cell membrane and bonded directly with DNA in the cytoplasm, leading to degradation, and/or (3) reacted to create Reactive Oxygen Species (ROS), as shown in Reaction 4.1, which degraded the cells through oxidative killing (Venis and Basu 2020). As such, the improved disinfection observed with Ag67ZnO33 in comparison to the other metal mixtures thus likely resulted from the higher concentration of silver present. With that said, further research into synergistic mechanics between these two nanoparticle species is needed.

4.3.2 SEM Imaging

SEM images were collected from *E. coli* cells without exposure to nanoparticles as well as after exposure to silver, zinc, and a combined solution (Figure 4.S1). Furthermore, all imaged

cells that were not treated by any nanoparticles remained entirely intact and presented strong visible contrast with the background surface (Figure 4.S1A). In Figures 4.S1B, C, and D, however, several observations can be made. The cells are elongated, twisted, and appear to have lower contrast with the background surface, indicating that the SEM electrons were actually able to penetrate cellular membrane due to increased permeability from lysis. Pits are also observed at edges on the *E. coli* cells indicating further damage to the bacteria. A notably greater number of pits were also observed in cells that were treated with the metallic mixture than those treated by either metal in isolation (see Figure 4.S1C inset), with more pits observed in cells treated by silver than by zinc, as expected.

4.3.3 Real Water Analysis

4.3.3.1 Nanoparticle Disinfection in Natural Waters

Though experimentation using synthetic water offers omniscient control of the parameters of interest, and therefore allows for improved elucidation of relationships between treatment variables, understanding how those same treatments translate under real water conditions with multi-modal and complex matrices is crucial. As such, nanoparticle disinfection in natural water drawn from the Ottawa River (Ontario, Canada) was also evaluated as a comparison to the synthetic water for the pH 7.4 condition. And as shown in Figure 4.6, all metallic nanoparticle bactericidal efficacy decreased when challenged by natural water compared to the synthetic water. It is important to note, however, that the silver-zinc combinations still outperformed the single component disinfection results. For instance, Ag1, ZnO1 and Ag67ZnO33 achieved LRVs after 300 minutes of -1.14±0.04, -0.47±0.06,

and -3.01±0.2 with the synthetic water conditions versus -0.55±0.08, -0.29±0.03 and -2.24±0.23 in the natural water, respectively ($p<0.01$). Note again that the Ag67ZnO33 combination in natural waters still outperformed silver alone in the synthetic water.

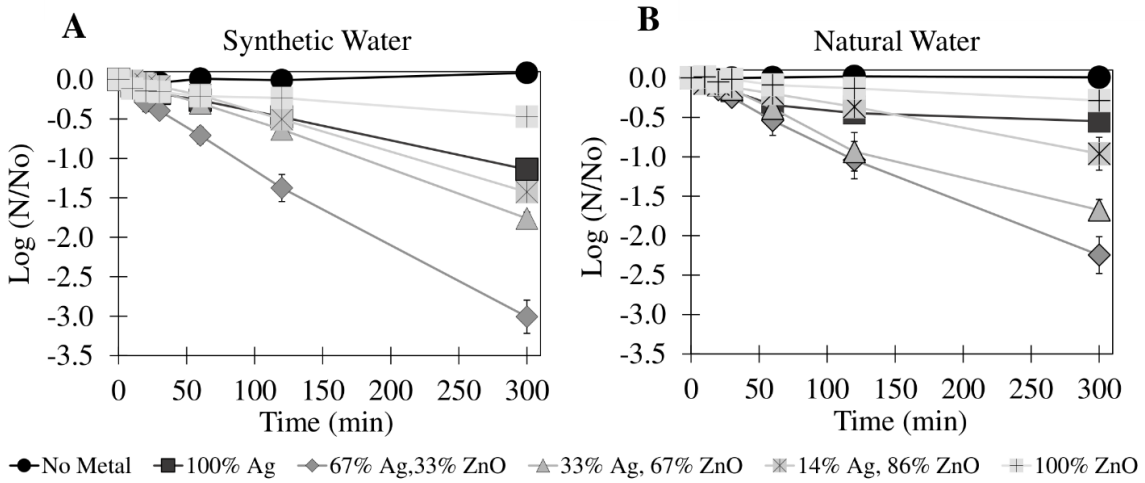
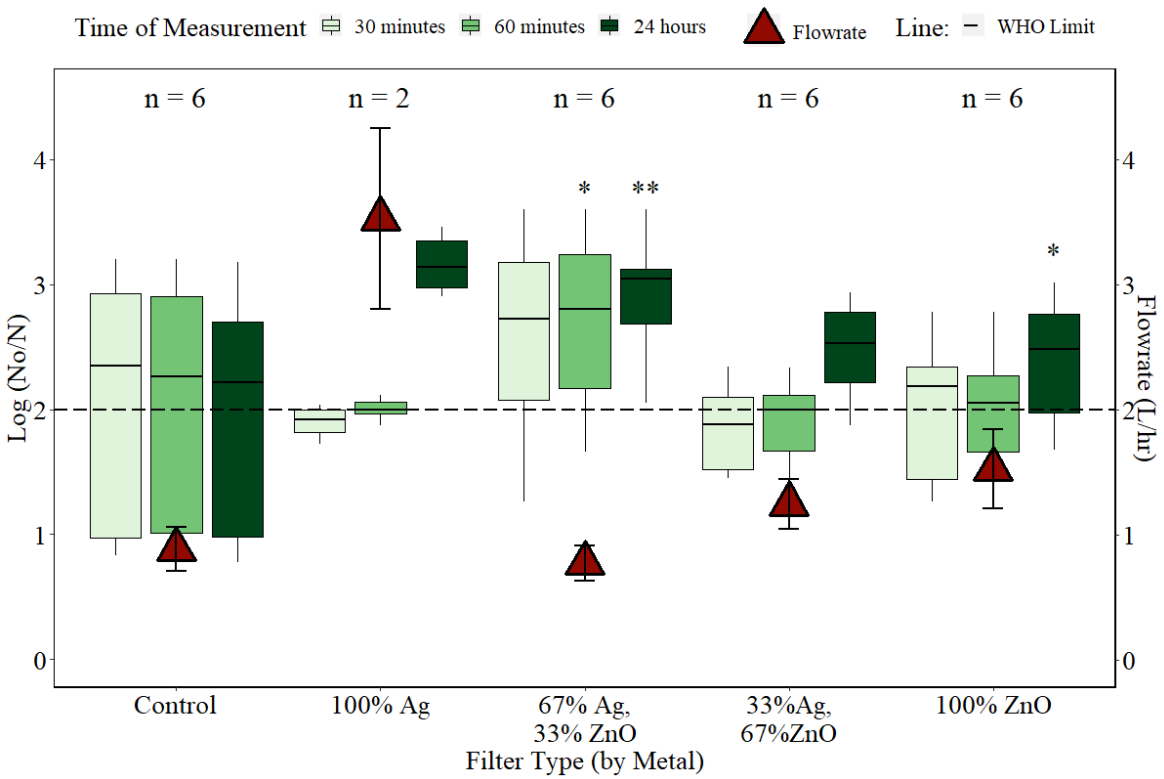


Figure 4.6 Disinfection achieved by each metal species and combination under synthetic (A) or natural (B) water conditions.

The observed reductions in the natural water may be attributed in significant part to the impact of natural organic matter (NOM) presence, which was approximately 8 mg/L as dissolved organic carbon (DOC) in this study. Previous research has demonstrated that NOM can coat the outside of nanoparticles and create a barrier between it and cell which inhibits interaction and reduces disinfection efficacy (Fauss, et al. 2014, Fabrega, et al. 2009). Attention to such water chemistry conditions is therefore important for application purposes when metals will commonly be exposed to organic matter. Nevertheless, these results importantly demonstrate that even in a complex (natural) water matrix, the combination of silver with zinc resulted in better disinfection than either metal species alone.

4.3.4 Ceramic Pot Filter Challenge Test

Results presented above clearly demonstrate that combined silver-zinc nanoparticle disinfection is robustly superior to that achieved by either single metal species across diverse water quality conditions. Investigation of how such observations translate to point-of-use water treatment applications is thus critically important for evaluating their implications in real-world applications. In these tests, samples were collected from the pot filtrate, meaning observed removals result from both filtration (size exclusion) and nanoparticle disinfection. As such, the control pots (with no metal nanoparticle addition) demonstrate that, as expected, the size exclusion is a key component to improving the water quality in this technology. As shown in Figure 4.7, however, results are far more variable with these filters than those with metals, and they straddle the WHO guideline of 2 Log Removal. This observation therefore suggests that the nanoparticles support immediate disinfection in the filtrate, as observed by a decreased variance in results; more testing is, however, recommended. Bacterial removals in the control filters also decreased slightly after 24 hours of storage when compared to immediately after filtration ($p = 0.13$), indicating a lack of continuous disinfection during storage. Conversely, all filters impregnated with nanoparticles achieved improved bacteria removal levels after 24 hours when compared with immediately after filtration, as well as when compared to the control ($p<0.01$). These results highlight that the nanoparticles predominantly impact bacteria removal after filtration once eluted into the receptacle, and that they importantly add value by helping maintain safe water quality in storage.



* 25% or more of samples achieved 100% bacteria removal
**50% or more of samples achieved 100% bacteria removal

Figure 4.7. Log Removal Values achieved by Ceramic Pot Filters impregnated by varying concentrations of silver and zinc oxide nanoparticles.

The filters containing 67% Ag and 33% ZnO were the only ones to significantly differ ($p<0.01$) from the no metal control filters after 60 minutes, achieving an LRV of 2.60 ± 0.6 compared with 2.10 ± 0.97 for the control filters. Further, more than 25% of evaluated samples for these pots achieved 100% removal after 60 minutes, increasing to more than 50% after 24 hours, meaning the achievable LRV in these filters was constrained by the influent *E. coli* concentration. As such, this trial supports the batch phase experimentation on utilizing a combined silver-zinc matrix to maintain and/or enhance disinfection of *E. coli* by ceramic filters. More experiments are therefore recommended on this topic, and

specifically in terms of varying metallic concentrations and methods of application (i.e., painting and submergence versus co-firing).

4.4 Conclusion

Advancements in water treatment practices are necessary if the world is to achieve the UN SDG 6 of providing safe drinking water for all people, and metallic nanoparticle disinfection offers an important opportunity for innovation. In the present work, the disinfection efficacy of combined and isolated zinc oxide and silver nanoparticles were evaluated in (1) batch experiments in synthetic and natural water and in (2) ceramic pot filters challenged by natural water. This research resulted in multiple significant findings:

- AgNP and ZnO demonstrated a synergistic relationship between the two species across pH conditions ranging from 6.4-8.4, as well as variable DO conditions.
- *E. coli* removal was fairly robust to changes in pH over the respective nanoparticle solutions tested.
- DO drastically impacted both the achieved disinfection and the associated kinetic rates, proving its significance as a factor in nanoparticle disinfection and highlighting a limitation of this water treatment solution if exposed to low DO waters.
- The best performing Ag/ZnO combo was a 67/33 mass ratio, which demonstrated a maximum average LRV of 3.18 compared to 1.4 for silver and 0.66 for zinc alone.
- Ceramic filters with silver-zinc combinations performed as well as, or in the case of filters with Ag/ZnO ratio of 67/33, superior to, filters with only silver, suggesting batch-scale experimental results translate to technologic application. The inclusion

of nanoparticles clearly indicated a beneficial impact on maintaining and improving water quality under storage conditions of 24 hours.

The clear improvement in bactericidal efficacy of silver and zinc nanoparticles when added in combination compared with when used in isolation, both in batch and pot phases, highlights a tremendous opportunity for innovative technological advancement in the field where technologies may be improved in terms of both effectiveness and affordability.

4.5 Acknowledgements

We would like to thank Argenol Laboratories for freely providing the silver nanoparticles in this research. We would also like to thank Wine to Water (non-governmental organization) for their support and collaboration, as well as all members of the Basu Research Group. This work was supported by the Natural Sciences and Engineering Research Council (NSERC), Canada, and Carleton University. This article is dedicated to Messiaki "Kim" Kimrei (d. 2020), former operator and owner of Safe Water Ceramics of East Africa (now W2WEA). His enthusiasm to explore change and improve the filters was a testament to his desire to bring safe water to his fellow Tanzanians.

Chapter 5: Elution and Disinfection of Silver and Zinc Nanoparticles in Co-Fired Ceramic Water Filters

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Submitted to Water Research

Abstract:

Ceramic water filters (CWFs) are decentralized water treatment technologies commonly used in resource-restricted geographies. Inclusion of silver nanoparticles (AgNP) assists with disinfection but can substantially increase costs. This research investigates AgNP supplementation with zinc oxide (ZnO) as a low-cost bactericide alternative. CWF disks were impregnated with varying AgNP and/or ZnO concentrations and challenged against *Escherichia coli*. Effluent bacteria were enumerated and monitored over 72 hours while eluted metal concentrations were measured and reproduced under batch conditions (0-20 ppb Ag, 0-800 ppb Zn). In latter experiments, bacterial regrowth was observed after 72 hours when Ag and Zn were below 10 ppb and 160 ppb, respectively. Meanwhile, 1 ppb Ag and 800 ppb Zn maintained complete disinfection for 72 hours. Increased Zn concentrations thus reduced Ag required to maintain disinfection over time. Pot-equivalent elution from CWF disks was estimated as 0-50 ppb Ag and 0-1200 ppb Zn, the respective amounts of which impacted

bactericide during storage. The average pot-equivalent elution estimate from CWFs impregnated with only Ag was 40 ppb Ag and 460 ppb Zn, which achieved a Log Removal Value (LRV) of 4.9 after 24 hours. Those with 33% Ag and 67% ZnO released an average pot-equivalent estimate of 20 ppb Ag and 515 ppb Zn and achieved an LRV of 4.8. Ag-Zn disinfection synergy was thus observed in the CWF phase, though Zn impregnation did not directly translate to elution. The presence of a background Zn concentration is suggested. Clay elemental composition may impact filter performance more than previously considered.

Keywords: Household water treatment; Metal nanoparticles; Metal release; Synergistic disinfection; Water and sanitation.

Highlights

- Ag and Zn synergistically disinfect *E. coli* in ppb-level concentrations
- Bacteria regrew over 72 hours if less than 10 ppb Ag and 160 ppb Zn present
- Ag supplementation with Zn can improve or maintain CWF disinfection
- Background Zn concentrations observed in CWF clay, impacting disinfection
- Zn impregnation did not directly translate to CWF elution via co-firing

5.1 Introduction

Rural and low-income populations in the global South are overwhelmingly those most disproportionately burdened by a lack of access to safe drinking water (World Bank 2021). Individuals within these areas are thus particularly subject to risk of associated physical health, mental health, social, and economic challenges (GBD Risk Factor Collaborators 2020). Climate change also threatens to exacerbate existing inequities by increasing water contamination and water-borne disease transmission over time through a combination of increased frequency and erraticism of storm events and decreased dilution capacity during droughts or periods with high temperatures (Bates, et al. 2008). The need for expanded affordable, accessible, and safely managed drinking water services is therefore urgent. Achieving progress within the areas most at risk has, however, proven challenging. That is, limited sectoral financial capacity (OECD 2020), vast and sparsely populated geographies, limited electricity supplies (World Bank 2021), programmatic challenges (Martin, et al. 2018), and various other factors continue to inhibit the proliferation of utility-scale drinking water services (Ray and Smith 2021). Alternatives to centralized water treatment engineering practices are thus required in the immediate term.

Point-of-use water treatment solutions (POUWTS) have responsively gained significant research attention (Lockwood, et al. 2010). POUWTS are small-scale water treatment technologies that can be used at the home and/or institutional level (e.g., schools, health clinics). Ceramic Water Filters (CWFs) are a particularly noteworthy POUWTS, widely

reported to exceed international guidelines for drinking water treatment efficacy (Oyanedel-Craver and Smith 2008, WHO 2011) while remaining among the least expensive and most preferable solutions to users (Santos, Pagsuyoin and Latayan 2016, Venis and Basu 2020). Yet, despite their comparative affordability, cost has been identified as a key limitation to widespread adoption (Burt, et al. 2017, Luoto, et al. 2012).

CWFs are typically manufactured by combining clay, a firing material (e.g., sawdust, rice husks), and water together into a homogenous mixture, which once pressed into a desired shape and dried, is fired in a kiln. The firing process burns away the firing material, producing a porous network throughout the filter body to remove contaminants by size exclusion (Venis and Basu 2020). Additionally, silver nanoparticles (AgNPs) are commonly impregnated into filters by one of two techniques: (1) adding the AgNPs into the mixture before firing (A.K.A, co-firing method), or (2) painting/submerging the CWF with an AgNP solution after firing (A.K.A, paint-on/submergence method). Ag is then released into the effluent during filtration, providing additional treatment capacity by disinfecting microbiological contaminants that pass through the matrix while maintaining a residual disinfection efficacy over time. However, silver's cost, procurement, and potential long-term health impacts that have raised concerns (CMWG 2010). Namely, AgNPs are commonly imported by CWF manufacturers in lower-income countries and can range in price between $4 and $20 USD/g, significantly influencing filter affordability (Sigma-Aldrich 2022). Consumption of more than 10 g AgNPs over a 70-year lifespan may also contribute to conditions such as Argyria or DNA damage (Fewtrell, Majuru and Hunter 2017, WHO 2011). CWF manufacturers are therefore forced to balance cost, health, and disinfection

efficacy when using silver in their products. Investigating Ag replacement or supplementation with alternative metallic nanoparticles (MNPs) may offer opportunity to mitigate these concerns.

Research into alternatively impregnated CWFs remains quite limited. However, some studies have evaluated MNPs like cupric oxide (CuO) with promising results (Lucier, Dickson-Anderson and Schuster-Wallace 2017). For example, Lucier et al. (2017) found that co-fired CWFs with no MNPs, 100% AgNP, 100% CuO, and a 2:1 CuO:AgNP mixture achieved average Log Removal Values (LRV; Log (C_o/C)) of 4.3, 4.5, 4.4, and 6.2, respectively, after 24 hours of storage post-filtration. The combined MNP impregnation thus led to a 2-fold improvement in bacteria removal, though eluted MNP concentrations were not assessed. On a batch-scale, Garza-Cervantes et al. (2017) also reported Ag-Cu disinfection synergy with higher LRVs among combined MNPs than either in isolation. The evaluated concentrations were, however, specific to medical applications and thus far exceeded international drinking water guidelines (WHO 2011, Venis and Basu 2020). In any case, evidence exists for Ag supplementation with CuO for advanced disinfection. Yet, its relatively high cost ($4-8 USD/g CuO) suggests its inclusion may not overcome the core concern of reducing filter price (Sigma-Aldrich 2022).

Another MNP that has garnered recent interest is Zinc Oxide (ZnO), which is available at a significantly lower cost than silver and has demonstrated efficacy as a treatment to diarrheal illness (Venis and Basu 2021, Sigma-Aldrich 2022, Malik, et al. 2013). For instance, Huang et al. (2018) found that ceramic disk filters painted with ZnO solutions of 8 and 41 mg/L achieved LRVs of 1.4 and 1.5, respectively, showing no significant effect immediately after

filtration. Eluted concentrations were not assessed. Meanwhile, when combined with silver at the batch scale for one hour, Garza-Cervantes et al. (2017) reported 32.7 mg/L of Zn^{2+} and 3.24 mg/L Ag^{+} achieved an LRV of 3.8 against *E. coli*. The same concentrations of Ag^{+} and Zn^{2+} alone achieved LRVs of 1.0 and -0.5 (i.e., growth), respectively. Similarly, Venis and Basu (2021) showed clear Ag-Zn synergy when a combined 1 mg/L of AgNPs with ZnO achieved superior disinfection over 5 hours of mixing than either MNP at the same concentration in isolation. Silver supplementation with ZnO is thus a particularly encouraging approach to potentially reducing required Ag concentrations for CWF impregnation.

With that said, no studies are known to have enumerated Zn elution from a CWF, and available batch- or adsorption-scale studies have used concentrations that far exceed those expected (Garza-Cervantes, et al. 2017, Motshekga, et al. 2015). That is, silver is typically released by painted/submerged filters in concentrations ranging between 0.02-2 mg/L, while 0.0005-0.15 mg/L is typically released by filters impregnated using the co-firing method; note, too, that far more research has investigated the former than the latter, highlighting an area in need of further investigation (Venis and Basu 2020). MNPs released by CWFs can therefore be orders of magnitude lower than the concentrations evaluated in batch-scale research within literature to date. The specific disinfection impacts of MNPs, and particularly ZnO, at such low levels is thus largely unknown. Put another way, the translation of impregnated metal concentration to subsequent bacterial disinfection within CWFs after filtration (during storage) is not elucidated. Further, the impacts of MNPs on disinfection during storage after CWF filtration largely lack investigation in general. That is, van der

Laan et al. (2014) saw a significant improvement in LRV between effluent CWF samples taken <5 minutes and 660 minutes after filtration. Yet, few additional studies have evaluated longer storage times (Venis and Basu 2021, Lucier, Dickson-Anderson and Schuster-Wallace 2017), and none are known to have studied eluted MNP impacts over more than 24 hours. As effluents commonly sit in a CWF receptacle for days at a time, understanding associated effects during this stage is critical to establishing appropriate filter usage and management protocols. Additional exploration of Ag-ZnO impregnation into filters, as well as its subsequent effects over long storage times, is thus required to identify best practices for maximizing performance while minimizing cost.

This study details the influence of ZnO as an AgNP supplement within CWF water treatment performance. Specifically, the removal of *E. coli* by small-scale CWF disks and a full-scale CWF pot during both filtration and storage phases is evaluated and correlated with enumerated concentrations of released metals. Eluted concentrations from disks are also scaled to full CWF pot-equivalencies to facilitate a more representative comparison. These equivalencies are then applied to batch-scale disinfection experiments in challenge water containing two levels of bacterial contamination that match observed filtrate concentrations. The objective of this study is to demonstrate how MNP impregnation translates to associated CWF disinfection and highlight the value offered by the inclusion of ZnO as an AgNP supplement. Additionally, the research aims to illustrates the respective roles of filtration and impregnated-metal disinfection in CWF water treatment, providing important insights into appropriate usage and storage protocols after implementation.

5.2 Methodology

5.2.1 Ceramic Water Filter Production

Fifteen ceramic water filter disks (CWF_{DISKS}) and one ceramic water filter pot (CWF_{POT}) were evaluated in this research, which were manufactured at the Wine to Water East Africa facility in Arusha, Tanzania in 2019 and 2015, respectively. Please see (Venis and Basu 2021) for a detailed description of the production process.

Summarized briefly, locally sourced clay and sawdust were first combined in an industrial cement mixer until visibly homogeneous. AgNPs (Argenol Laboratories) and/or ZnO (Sigma Aldrich) were then dissolved in mass ratios of (1) 100% Ag/0% ZnO (2) 67% Ag/33% ZnO, (3) 33% Ag/67% ZnO, and (4) 0% Ag/100% ZnO into water to reach a 7.5 mg/kg total metal concentration (approx. 1 mg per disk). Metal impregnation thus followed the co-firing method. Unimpregnated (no metal addition) filter disks were also tested as a control case. The wet mixtures were then kneaded and pressed into a 2.5 cm thick container from which four disks were cut to 8 cm diameters. Filters were dried and subsequently fired in a kiln increasing in temperature to $960^{O}C$ over 12 hours and cooled to room temperatures over 12 hours thereafter. All filters were comprised of the same clay and sawdust ratios (46% clay to 54% sawdust by volume) and the same sawdust particle sizes (nominal Ø: 375µm) to obtain consistency in filtration performance. After firing, the disks were shaved with a course file to 2.2 cm thicknesses and 7.5 cm diameters to fit within the testing unit and match the CWF_{POT} wall thickness.

The filter pot was manufactured under a similar procedure with minor variations. Namely, only AgNPs were used for impregnation, as this filter represents the product currently sold and distributed by the manufacturer. The wet mixture was also pressed into the pot shape with a manual press and mold to obtain a rounded/paraboloid bottom with a height of 26 cm and rim diameter of 27 cm, as opposed to a flat container used with the disks. This filter was provided by the manufacturers in 2015 and was not specifically produced for this research.

5.2.2 Disk Filter Testing

5.2.2.1 Test Preparation

Before testing, each CWF_{DISK} (Figure 5.1(1)) was brushed with a hard bristle and gently washed with distilled water to remove any surface grime (Figure 5.1(2)). It was then submerged in boiled water with a measured dissolved oxygen (DO) concentration of <0.5 mg/L for 30-60 minutes or until no bubbles could be visually observed along the filter surface or within the water (Figure 5.1(3)). After submergence, the CWF_{DISK} was removed from the water and placed in a $65^{O}C$ oven for a minimum of 48 hours (Figure 5.1 (4)), or until the measured mass was equal to the dry mass measured before testing began. Once dried, filters were wrapped tightly in Teflon Tape (Figure 5.1(5)) to encourage one-dimensional flow. The wrapped disk was then placed in a holding unit and sealed at the top and bottom with food grade silicon glue and left to cure for a minimum of 12 hours (Figure 5.1 (6-8)). At the time of testing, the CWF_{DISK} within the holding unit was screwed into a threaded PVC pipe until water-tight, which was filled to 25cm of head (Figure 5.1(9)).

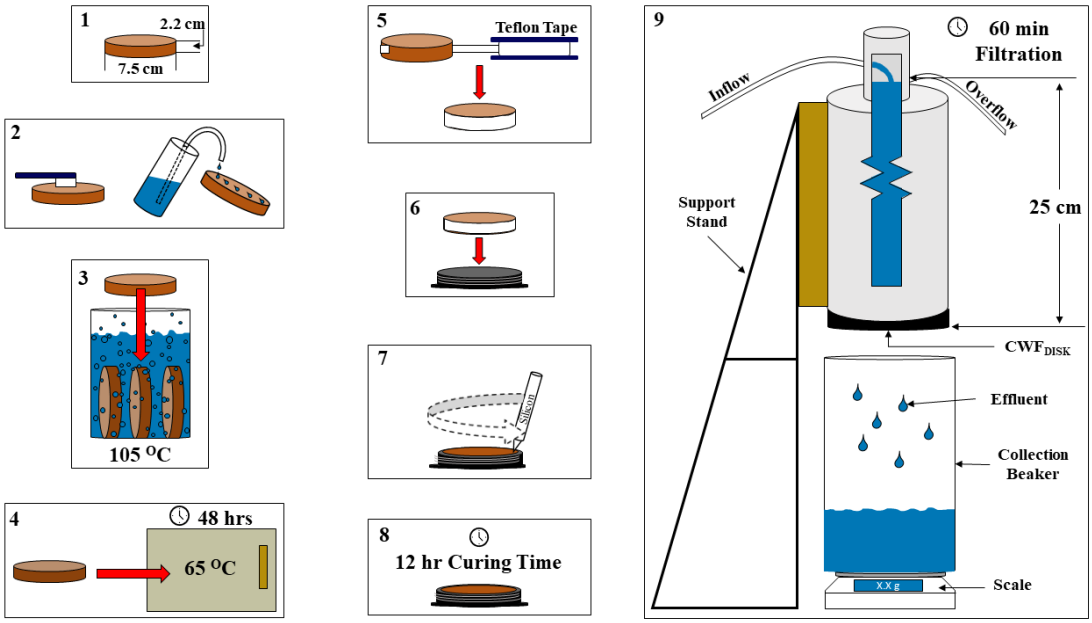

Figure 5.1 Schematic of CWF$_{DISK}$ preparation procedure

5.2.2.2 Microbial Culturing and Enumeration

K-12 Migula (ATCC® 29947) Castellani and Chalmers *E. coli* was purchased from Cedarlane laboratories as a dry pellet, which was rehydrated with 1 mL of No. 3 Nutrient Broth and centrifuged for 10 s. 200 µL of *E. coli* solution was injected into 9800 µL of broth and incubated for 1 hour at 37 ^{O}C. 1 mL of sterile, pure glycerol was then added to the stock solution, which was incubated again for 3 hours. The solution was then divided into 200 µL aliquots and stored at -20 ^{O}C. For testing purposes, frozen stock was thawed and 100 µL was added to 9900 µL of No. 3 Nutrient Broth (0.1% solution), which was incubated at 37 ^{O}C for 19 hours until the midpoint of the stationary growth phase; a 10^8 CFU/mL culture was resultingly achieved.

Bacterial concentrations were measured as per USEPA Method 1604 with the membrane filtration method (USEPA 2002). 1 mL samples were drawn from influent and effluent

vessels, which were appropriately serially diluted with a buffer solution (0.5% $MgCl_2$ and 0.125% KH_2PO_4) until a measurable concentration was reached. Diluted samples were then spread over a 0.45 µm filter paper (25 cm) and vacuum filtered. Papers were placed upon MI agar (Thomas Scientific) nutrient pads and incubated at 37 ^{O}C for 24 hours. Colonies were counted manually.

5.2.2.3 Influent Water Quality

CWFs were challenged by soft water of low alkalinity, as per Table 7 in (USEPA 2002). Influent water was injected with dissolved organics and micronutrients, as well as with bacterial stock to produce a concentration of approximately 10^5 CFU/mL. pH was adjusted with 1N solutions of HCl (acid) and NaOH (base). All water quality parameters were measured from the influent water at the beginning of each test and are detailed in Table 5.1.

Table 5.1 Water Quality of Challenge Water

Water Quality Parameter	Value (±SD)
pH	7.4 ± 0.05
ORP (mV)*	-18 ± 2
Dissolved Oxygen (mg/L)	8.70 ± 0.5
Temperature (^{O}C)	21.5 ± 1.0
Alkalinity (mg/L $CaCO_3$)	30 ± 2
Hardness (mg/L $CaCO_3$)	23 ± 2
*TOC** (mg/L)*	6.1 ± 0.2
E. coli (x10^5 CFU/mL)	3.32 ± 1.00
Ion Concentrations (mM)	
Ca^+	0.11 ± 0.01
Na^+	0.75 ± 0.02
Mg^{2+}	0.12 ± 0.01
K^+	0.01 ± 0.002

*ORP = Oxidation Reduction Potential
**TOC = Total Organic Carbon

5.2.2.4 Testing Procedure

After the desired head (25 cm) was reached, filters were left until one pore volume (approx. 35 mL of water) had passed through the matrix. Influent bacterial concentrations were then measured, and effluent measurements were taken after 30 and 60 minutes of filtration. Flowrate was calculated from the mass of filtrate collected in that time. After 60 minutes, collected effluent was transferred to a batch reactor and mixed constantly for 72 hours at 50 rpm with a flat blade propellor under dark conditions. The same water quality parameters were then measured at 24-, 48-, and 72-hour timepoints. *E. coli* was measured in duplicate at each time point. Three CWF_{DISKS} of each filter type were each tested twice (i.e., n = 12). The CWF_{POT} was tested twice with duplicate sampling (i.e., n = 4). Relevant statistical tables may be found in Appendix F.

All materials were cleaned with enzymatic detergent at the end of the experiment and rinsed thoroughly with distilled water. All autoclavable materials were treated thusly for at least 15 minutes at 121 ^{O}C and subsequently wrapped in aluminum foil to prevent recontamination before reuse.

5.2.2.5 Eluted Metal Enumeration

Zinc and silver concentrations were enumerated using inductively coupled plasma mass spectrometry (ICP-MS) by the Geochemistry and ICP Laboratory at the University of Ottawa in Ottawa, Ontario, Canada. 9800 µL samples were drawn from the filter effluent collection vessel after 30 and 60 minutes of filtration, which were acidified with 200 µL of pure nitric acid to create a 2% solution (volumes added isometrically). Samples were then labelled with their CWF name and date, wrapped in aluminum foil to prevent light penetration, and stored

at 4 ^{O}C for a maximum of 2 weeks before enumeration. Note that only total metal concentration (i.e., no disaggregation between zero-valent and ionic oxidation states) is measured under this ICP-MS procedure as the acidification process necessarily converts all metals to their ionic state. Concentrations are reported in parts per billion (ppb). Relevant statistical tables may also be found in Appendix F.

5.2.2.6 Disk-to-Pot Filter Scaling

Measured metal elution values were scaled with Equation 5.1 to predict eluted concentrations as if it were from a CWF_{POT} of the same size as that tested.

$$ME_P\left(\frac{\mu g}{L}\right) = ME_D\left(\frac{\mu g}{L}\right) \times \frac{SA_P\,(cm^2)}{SA_D\,(cm^2)} \qquad [5.1]$$

Where ME is metal eluted, SA is surface area, subscript P is for pot and subscript D is for disk. Pot surface area was further calculated from Equation 5.2, which is derived by integrating a revolution of Equation 5.3

$$SA_P(cm^2) = \int_0^{r_0} 2\pi r\sqrt{1+\left(\frac{r}{an}\right)^{\left(\frac{2}{n}\right)-2}}\,dr \qquad [5.2]$$

$$r = az^n \qquad [5.3]$$

where r is the filter radius, r_0 is the radius of the filter at the height of the water (13 cm), a (a > 0) and n (0 < n < 1) are fitting parameters calculated from the dimensions of the tested $CWF_{POT,}$ and z is the height of the filter from the base. SA_P was estimated to be approximately 1730 cm^2 and parameters a and n were estimated to be 2.89 and 0.49, respectively, with a standard error of 0.77%, shown in Figure 5.2.

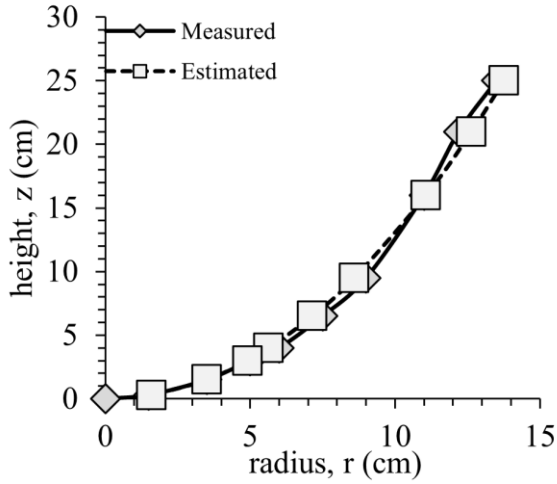

Figure 5.2 Estimated and measured CWF_{POT} height and radii.

In addition to pot-equivalent elution, constant head pot-equivalent flowrates (FR_P) were also calculated for filter comparisons as per Equation 5.4 from (Schweitzer, Cunningham and Mihelcic 2013).

$$FR_P\left(\frac{L}{hr}\right) = \frac{2\pi Ka}{t(n+1)(n+2)} \times z_0^{n+2} \qquad [5.4]$$

K is the disk's experimental hydraulic conductivity (cm/hr), t is the filter thickness (cm), and z_0 is the height of the water in the filter (cm). Any CWF_{DISK} tests with a pot-equivalent flowrate of higher than 4.5 L/hr were excluded from the analysis, as per the criteria used for determining if a CWF_{POT} may be sold by the manufacturer.

5.2.3 Batch-Phase Experiments

Batch-phase experiments were conducted with the same water chemistry conditions as described in Table 5.1. In these tests, however, initial bacterial concentration was lowered to coincide with levels observed immediately after disk filtration, as detailed in Table 5.2.

Table 5.2. Initial bacterial concentrations in batch experiments

Test Type	Reference Concentration (CFU/mL)	Actual Concentration (CFU/mL)
Low	10^2	558 ± 63
High	10^3	4481 ± 643

At 10^2 CFU/mL, 1, 5, 10, and 20 ppb AgNPs were injected with 0, 40, 160, 800 ppb as Zn (0, 50, 200, and 1000 ppb as ZnO); a no-metal control was also evaluated. At 10^3 CFU/mL, the same tests were conducted, however 1 and 5 ppb Ag were not evaluated with 40 and 160 ppb Zn as no disinfection was observed at these concentrations when challenged prior by 10^2 CFU/mL (see Section 5.3.4). These MNP concentrations were chosen to approximately align with eluted values after scaling to pot-equivalencies with the procedure described in Section 5.2.2.6. More specifically, silver concentrations ranged from the lowest pot-equivalency concentration to near the highest measured concentration from the CWF_{POT}. Zinc concentrations were chosen as slightly above 1μM, the limit at which zinc begins exhibiting toxic effects on *E. coli* (Yamamoto and Ishihama 2005), as well as the 10th and 90th percentiles for which pot-equivalent zinc concentrations were estimated (See Section 5.3.4). Full characterization of Ag and Zn NPs used in this research can be found in (Venis and Basu 2021).

E. coli was enumerated before metal injection, as well as 5, 24, 48, and 72 hours thereafter. All samples were taken in duplicate, and all tests were run twice (i.e., n = 4). Note that these experiments were designed as a quasi-simulated investigation of exclusively metal-based disinfection by a CWF_{POT} during storage. Please also note that the use of K-12 *E. coli* herein is an important study limitation as cells of this strain may be more susceptible to disinfection

impacts than others typically found in a natural environment. Additional research investigating alternative strains and/or species is recommended

5.3 Results and Discussion

5.3.1 Disk Filtration and Storage

Shown in Figure 5.3 is the achieved LRVs (Log C_o/C) from CWF_{DISKS} and the CWF_{POT} immediately after filtration (blue bars), as well as after storage (orange bars). Please note that all 100% ZnO filters were omitted from the present analysis because they failed to meet the flowrate inclusion criteria detailed in Section 5.2.2.6.

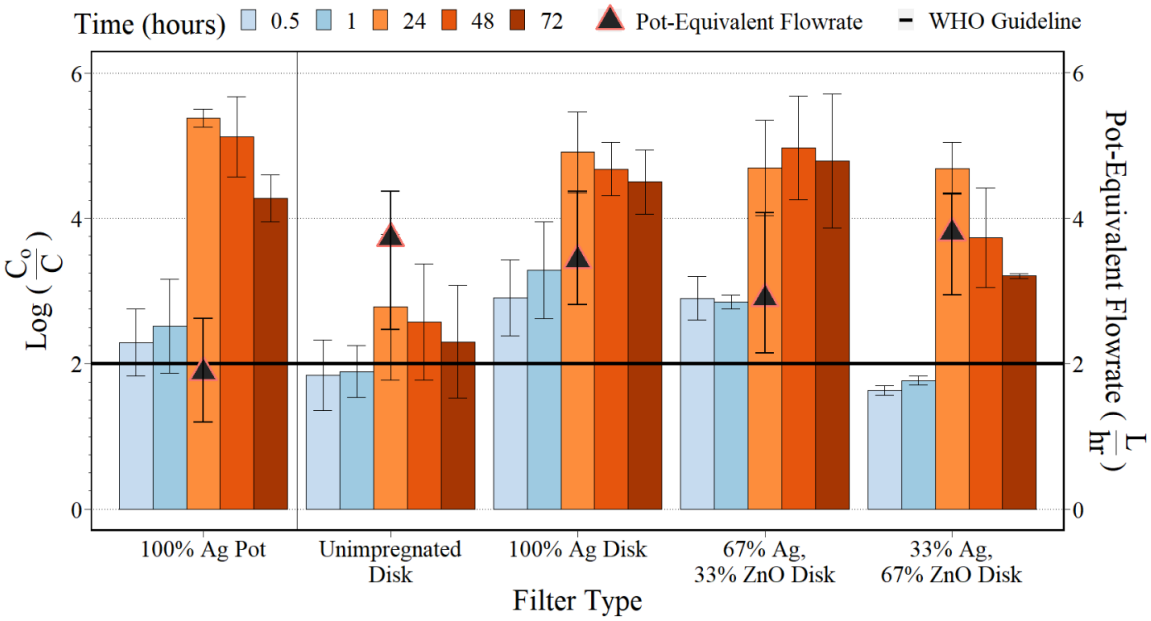

Figure 5.3. Log Removal Values achieved by CWF_{POT} and CWF_{DISKS} containing varying concentrations of AgNPs and ZnO immediately after filtration (blue), and during storage (orange).

Size exclusion during filtration (blue) evidently contributed significantly to overall bacterial removal in both pot and disk filters, highlighting its importance to CWF function. There were also no statistical differences observed between samples taken after 30 and 60 minutes of

filtration within a given filter type ($p>0.05$). The 100% Ag/0% ZnO and 67% Ag/33% ZnO CWF_{DISKS}, did, however, achieve slightly larger LRVs at 30- and 60-minute samples than all other filters (2.8-3.3 vs. 1.8-2.5; $p<0.05$). Also, the unimpregnated and 33% Ag/67% ZnO filters were the only types to not achieve average LRVs >2, the WHO guideline for the specific context of using household water filters (see Table 7.8 in (WHO 2011)). As such, though these latter filters were also those with the highest flowrates – a performance metric highly correlated with size exclusion efficacy (CMWG 2010) – the lower initial LRVs may indicate some, albeit minor, level of disinfection contribution from the MNPs.

Post filtration, the LRVs for all filters increased after 24 hours of storage ($p<0.01$), with metal-impregnated filters observing a significantly larger increase than the unimpregnated control. More specifically, CWF_{DISKS} with 100% Ag/0% ZnO, 67% Ag/33% ZnO, 33% Ag/67% ZnO achieved LRVs of 3.3 ± 0.66, 2.8 ± 0.1, and 1.8 ± 0.1 after 60 minutes, which increased to 4.9 ± 0.6, 4.7 ± 0.6, and 4.7 ± 0.4 after 24 hours, respectively. The potential to replace some Ag with ZnO is thus demonstrated. Further, while CWF_{DISKS} with 100% Ag/0% ZnO and 67% Ag/33% ZnO maintained similar disinfection levels over the 72-hour evaluation period, those with 33% Ag/67% ZnO decreased in LRV (i.e., bacterial regrowth was observed). Conversely, the unimpregnated CWF_{DISKS} achieved an LRV of 1.9 ± 0.4 post filtration, which then increased to 2.8 ± 1.0 after 24 hours and decreased to 2.3 ± 0.08 ($p<0.05$) after 72 hours. This initial improvement, though lower than all other filters ($p<0.01$), was still significant and may further be attributed to measured eluted zinc from the inherent clay material itself (see Section 5.3.2). These results highlight that the metallic

contribution to disinfection occurs primarily in the receptacle during storage (Venis and Basu 2021, van der Laan, et al. 2014), with marginal impacts observed during filtration.

5.3.2 Metal Elution

Both measured and predicted metal elution concentrations are detailed in Table 5.3. A first important observation is that the 100% Ag/0% ZnO CWF_{DISK} released higher concentrations of silver (40.6 ± 13.9 ppb) than the CWF_{POT} (16.2 ± 1.8 ppb) after the pot-equivalence conversion, suggesting higher MNP concentrations may be released by disks than pots per unit area. This difference is aligned with expectation as the gravitational force applied to the CWF_{DISK} was higher than that applied to the CWF_{POT} per unit area because of the one dimensional versus radial flow regimes. CWF_{DISKS} were thus likely subject to comparatively increased pore pressures, translating to greater metal release than the CWF_{POT}.

Although few studies have evaluated elution from co-fired filters, these results are also similar to those observed among other co-fired CWF_{DISKS} (Jackson and Smith 2018, Ren and Smith 2013) and CWF_{POTS} (Shepard, Lux and Oyanedel-Craver 2020, Venis and Basu 2020). That is, as detailed previously, 0.5 to 150 ppb Ag have been reported to elute from co-fired CWFs depending on the impregnation concentration. Meanwhile, painted/submerged filters having released Ag in concentrations as high as 11000 ppb (Venis and Basu 2020). This study therefore confirms that the co-firing method leads to significantly lower Ag releases than alternatives. All elution concentrations were also statistically similar between 30- and 60-minute samples within each filter type ($p>0.1$) and previous research has shown Ag elution from co-fired CWFs remains consistent over time (Ren and Smith 2013).

Comparatively, Ag elution from painted/submerged CWFs have been shown to change with the volume of water filtered (Jackson and Smith 2018, Oyanedel-Craver and Smith 2008). That said, this research did not evaluate elution beyond 60 minutes of filtration, meaning long-term trends in metal release from co-fired filters remains relatively unelucidated. Additional research investigating co-fired filter elution over longer filtration time periods is thus recommended.

The CWF_{POT} also released less zinc than its disk counterpart, though this difference was much larger than that of silver. Whereas the CWF_{POT} released an average of 1.61 ± 1.39 ppb Zn, the 100% Ag/0% ZnO CWF_{DISK} released 11.3 ± 4.2 ppb Zn, or 460.8 ± 173.9 ppb Zn when scaled to a pot equivalent. A background concentration of Zn thus evidently existed within the disk filters that did not exist in the pot, as confirmed by the measured Zn elution of the unimpregnated CWF_{DISK} (13.1 ± 10.0 ppb). This difference is hypothesized to have emerged because of differences in the time of manufacture. The CWF_{POT} was produced in 2015 with different clay and sawdust sources than those used for the CWF_{DISKS} in 2019. X-ray fluoresecence (XRF) results also indicated the presence of 140 $\mu g/cm^3$ of ZnO in the clay from CWF_{DISKS} produced in 2019 and no detectable zinc concentration in clay used for the CWF_{POT}, collected from the factory in 2015. Additionally, no zinc is reported in the composition of the non-filter materials (e.g., the silicon glue). As such, differences in the input materials' elemental compositions most likely resulted in the observed differences in eluted metal concentrations.

Table 5.3. Eluted zinc and silver concentrations as measured and as scaled to pot-equivalents

Filter Type	Silver Average	Silver Range	Zinc Average	Zinc Range
Measured Disk Elution (ug/L)				
100% Ag, 0% ZnO	1.00	0.51 - 1.40	11.33	5.84 - 15.32
67% Ag, 33% ZnO	0.59	0.11 - 1.29	17.20	6.11 - 29.20
33% Ag, 67% ZnO	0.40	0.04 - 1.26	11.88	5.31 - 18.13
Unimpregnated	0.05	0.04 - 0.06	10.06	3.77 - 26.15
Estimated Pot-equivalent Elution (ug/L)				
100% Ag, 0% ZnO	40.62	20.51 - 56.25	460.82	235.34 - 617.48
67% Ag, 33% ZnO	25.48	4.55 - 54.91	722.52	260.09 – 1208.74
33% Ag, 67% ZnO	17.58	1.58 - 55.15	516.94	216.67 – 793.64
Unimpregnated	2.05	1.65 – 2.48	544.33	156.11 - 1082.86
Measured Pot Elution (ug/L)				
100% Ag, 0% ZnO	16.17	14.07 - 17.86	1.61	0.62 - 3.84

Another critical note is that impregnated silver concentration translated fairly directly to observed elution, however the relationship between zinc impregnation and release was more variable. For instance, CWF_{DISKS} with 100% Ag/0% ZnO, 67% Ag/33% ZnO, and 33% Ag/67% ZnO released 40.6 ± 13.9, 25.5 ± 21.7, and 17.6 ± 23.3 ppb Ag as pot-equivalencies, respectively, demonstrating a clear trend of a 30-40% increases in eluted value with respective increases in impregnation. The same CWF_{DISKS}, however, released 460.8 ± 173.9, 722.5 ± 364.0, and 519.9 ± 216.6 ppb Zn as pot-equivalencies, respectively. An omnibus ANOVA further confirmed no differences in pot-equivalent Zn elution for any of the filter types, except for the CWF_{POT} that differed from all other filters ($p<0.01$). The amount of ZnO impregnated into the filters during fabrication (~30-60 μg/cm^3) therefore had no observable impact on the Zn release. The background zinc concentration (70-80% of total

present Zn) within the clay is moreover hypothesized to have overpowered that which was impregnated, suggesting the elemental composition of clay sources may play a more significant role in CWF disinfection than previously considered.

This lack of translation between Zn impregnation and elution further highlights a critical need for additional research. Namely, these observations highlight a key matter that CWF researchers have seldom studied in detail: how clay composition influences bacteria removal performance. For instance, Shepard et al. (2020) evaluated 13 different clay sources from various locations and found significant differences in biofilm formation on each surface, yet elemental composition was not enumerated nor was filtration evaluated. As such, while clay source appears particularly relevant to CWF treatment performance, research is required to identify the degree of variability that exists, how that variability impacts disinfection, and how it may be controlled to ensure CWF manufacturers produce high-quality filters regardless of geography.

A final important note is the high degree of variability that existed for both Ag and Zn release. Namely, variability was observed between individual CWF_{DISKS} of the same type, and between tests. Elution thus appears not only correlated with water quality and filter properties, but also with the initial conditions in which water passes through the filter matrix. That is, differences in flowrates, as shown in Figure 5.4, may have influenced the contact times between the water and the impregnated metals, impacting elution in turn. Specifically, pot-equivalent Ag and Zn concentrations were plotted against pot-equivalent flowrate for each individual CWF with the "plotly" package in R to produce the observed contours. The

highest flowrates are observed among the lowest measured elution concentrations, while slower flowing filters released higher metal concentrations.

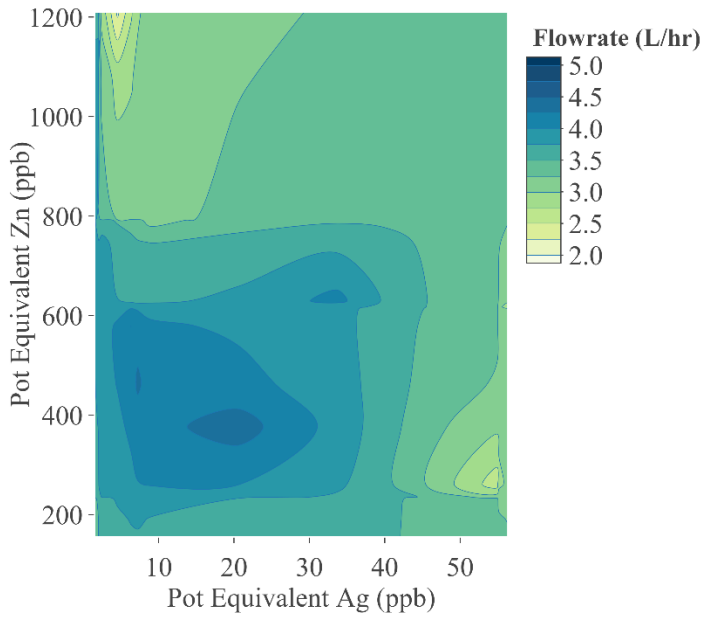

Figure 5.4. Contour Plot of Pot-equivalent Flowrate as a function of Pot-equivalent Elution of Ag and Zn. Each contour line represents a change of 0.25 L/hr.

5.3.3 Elution-Disinfection Interaction

How metal release contributed to disinfection is paramount. Contour plots were produced for LRV as a function of Ag and Zn and measured releases (Figure 5.5) from each filter. Overall, as was also observed in Figure 5.3, the LRV achieved at each time point generally decreases as storage time increases. The influence of metals is also evidently less significant after 1 hour of filtration (Figures 5.5A) relative to after storage (Figures 5.5B-D), with higher LRVs observed from higher combined concentrations of Ag and Zn.

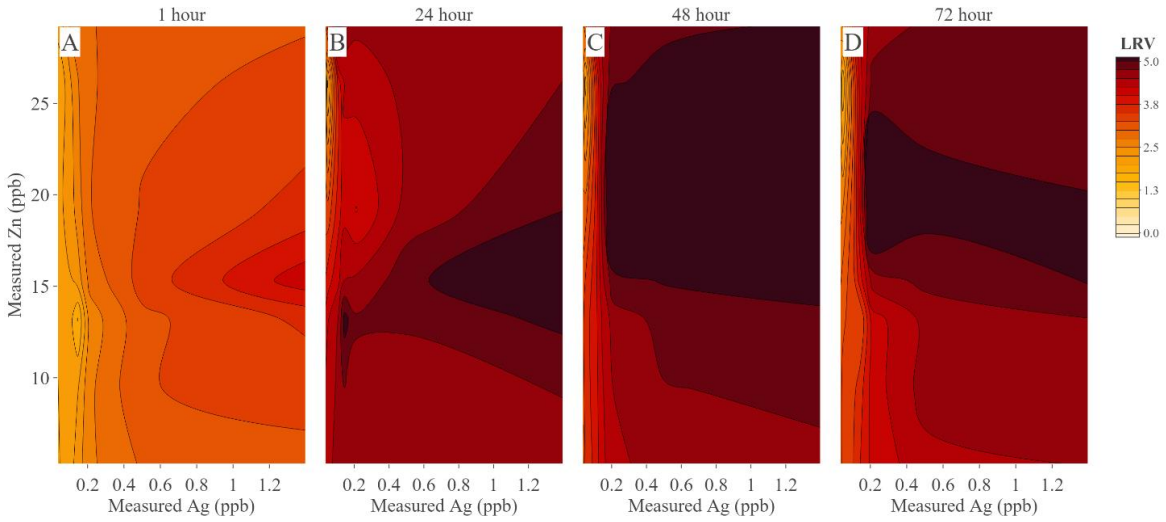

Figure 5.5 Contour plots with Log Removal Values (LRV) from disks according to measured eluted silver and zinc after (A) 1 hour of filtration, (B) 24 hours of storage, (C) 48 hours of storage, and (D) 72 hours of storage. Note that each contour line represents a change of 0.25 in LRV

After 24 hours of storage, LRV improves with increasing Ag and Zn concentration. For example, measurements of 0.5 ppb Ag and 9.4 ppb Zn achieved an LRV of 4.5, whereas 1.4 ppb Ag and 15.3 ppb Zn achieved an LRV of 5.6. After 48 and 72 hours, the influence of Zn then increased. A trend towards higher LRVs with higher Zn concentrations is observed in Figures 5.5C, and 5D. Specifically, whereas 0.15 ppb Ag and 13.2 ppb Zn achieved LRVs of 4.3 and 3.2 after 48 and 72 hours, respectively, 0.11 ppb Ag and 29.2 ppb Zn achieved LRVs of 4.3 and 4.0, respectively. Similarly, 0.21 ppb Ag and 19.1 ppb Zn achieved an LRV of 5.5 after 48 hours, which was maintained to 72 hours. Zn concentration thus demonstrably led to improved maintenance of achieved disinfection in the time prior.

5.3.4 Batch Experiments

As detailed in Section 5.2.3, metal concentrations evaluated in the batch phase were approximated derivatives of CWF_{DISK} elution observations after scaling to a pot-equivalency,

allowing for a more detailed evaluation of the exclusive contributions to disinfection by Ag and Zn species. 1, 5, 10, and 20 ppb Ag were tested in combination with 0, 40, 160, and 800 ppb Zn (0, 50, 200, and 1000 ppb as ZnO). As may be seen from Figure 5.6, several observations from the filter phase translated to the batch phase, albeit with some outliers. A first note is that achieved disinfection for all Ag levels increased with increasing ZnO concentrations. For instance, after 24 hours, bacterial growth was observed when 5 ppb Ag was combined with Zn in concentrations up to 160 ppb in water with 10^2 CFU/mL. When combined with 800 ppb Zn, however, an LRV (Log C/C_O) of -2.67 ± 0.1 (disinfection) was measured. Similarly, Zn's contribution to disinfection appeared more effectual over time and with larger Ag concentrations. Namely, at 24 hours, 20 ppb Ag achieved LRVs of -1.25 ± 0.1, -0.76 ± 0.1, -2.44 ± 0.3, and -2.70 ± 0.01 with no Zn, 40 ppb Zn, 160 ppb Zn, and 800 ppb Zn against 10^2 CFU/mL, respectively. At 72 hours, the same metal combinations achieved LRVs of -0.11 ± 0.1, +0.11 ± 0.03 (growth), -2.79 ± 0.04, and -2.70 ± 0.01, respectively. Bacterial regrowth was thus observed when Ag was challenged alone or with 40 ppb Zn, and LRV either remained the same or improved when in combination with 160 ppb Zn or more.

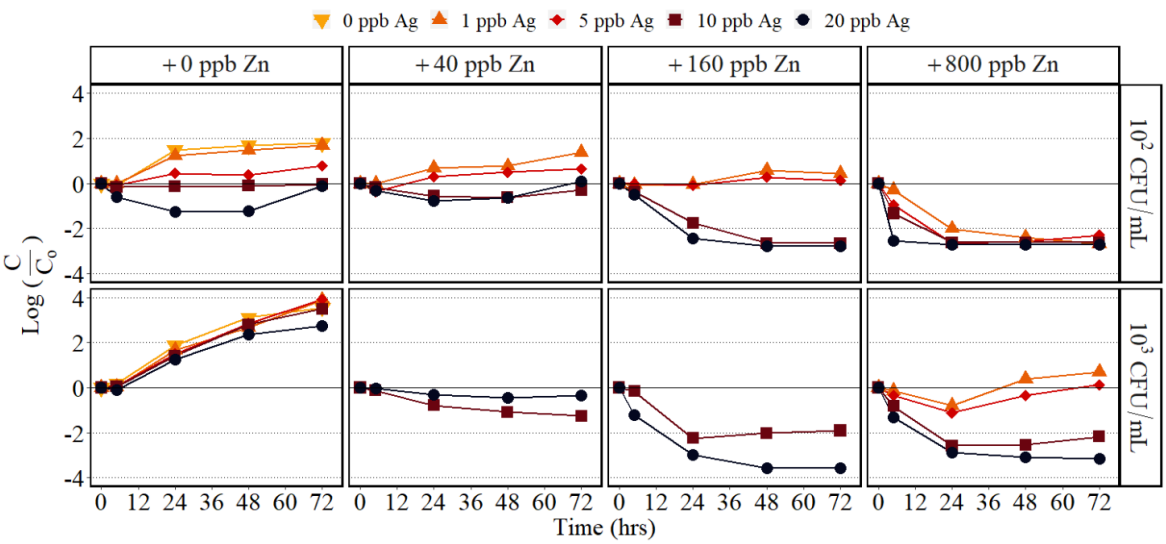

Figure 5.6 Log Removal Values achieved by silver and zinc combinations over 72 hours in water with 10^2 CFU/mL (top row) and 10^3 CFU/mL (bottom row)

While CWFs are designed to have effluent bacterial concentrations of less then 10^2 CFU/mL, studies have shown such objectives are commonly unmet (Meierhofer, et al. 2018). It is thus important to understand the influence of nanoparticle disinfection under more severe bacterial exposures. For instance, whereas 10 ppb Ag alone reached an LRV of -0.10 ± 0.14 after 48 hours when challenged by water containing 10^2 CFU/mL, log growth of +2.80 ± 0.08 was observed in water with 10^3 CFU/mL. Similarly, 1 ppb Ag combined with 800 ppb Zn achieved an LRV of -2.58 ± 0.18 (disinfection) in water with 10^2 CFU/mL after 48 hours compared with a log growth of +0.38 ± 0.02 in water with 10^3 CFU/mL. Meanwhile, 10 ppb Ag and 800 ppb Zn achieved an LRV of -2.61 ± 0.03 in water with 10^2 CFU/mL after 48 hours compared with -2.54 ± 0.14 in water with 10^3 CFU/mL. Higher concentrations of either Ag or Zn therefore did not protect against bacterial growth in water with a higher initial bacterial concentration unless combined, suggesting both metals are needed in sufficient quantities for consistent results across potentially variable water qualities.

5.3.5 Batch-CWF Comparisons

The correlation between batch and CWF disinfection may be observed in Figure 5.7. Specifically, Figures 5.7A-C represent the storage disinfection achieved by both CWF_{DISKS} and the CWF_{POT}, with elution values for CWF_{DISKS} scaled to pot-equivalencies. In other words, LRVs are calculated as the difference between effluent bacterial concentrations after one hour of filtration to those measured during the storage phase. Figures 5.7D-F detail the disinfection achieved during the batch phase, disaggregated by influent bacterial concentration.

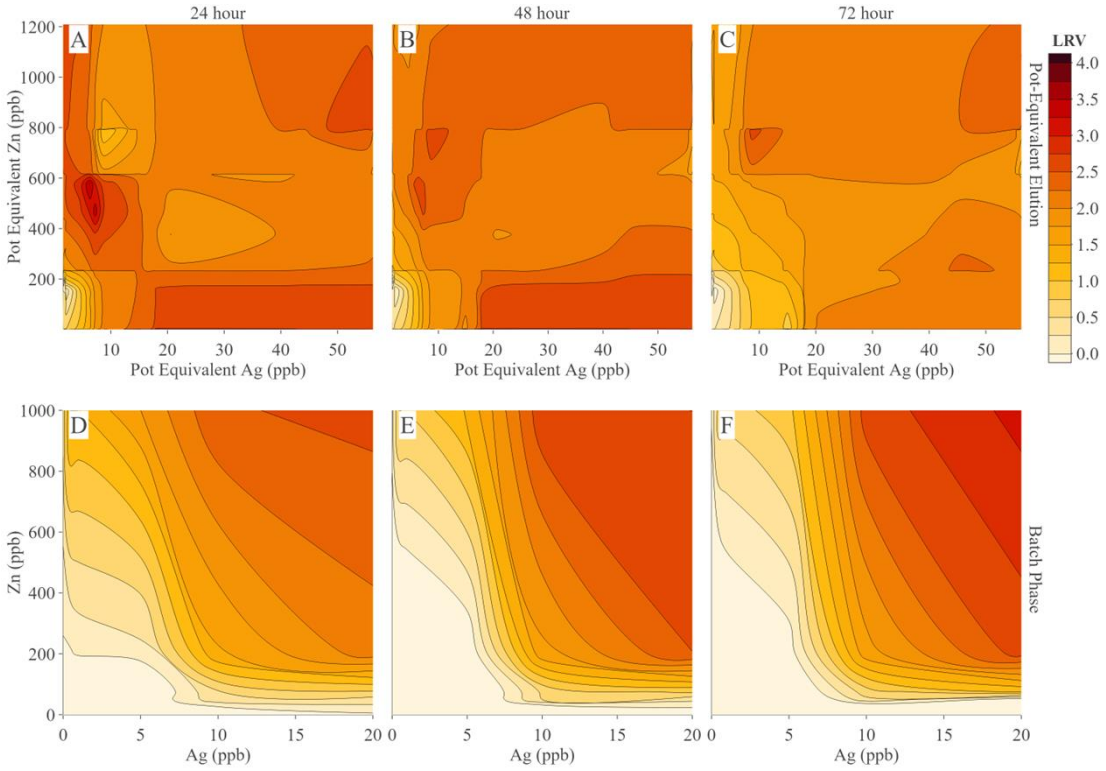

Figure 5.7. Contour plots displaying LRVs achieved CWFs per pot equivalent MNP release after (A) 1 hour (i.e., post-filtration), (B) 24 hours, (C) 48 hours, (D) 72 hours, and LRVs achieved by MNPs during batch-phase experiments without controlling for initial bacterial concentration after (E) 5 hours, (F) 24 hours, (G) 48 hours, (H) 72 hours.

A first notable observation is the generally decreasing effectiveness of all metals with storage time in both filter and batch phases. That is, some bacterial regrowth was observed in all cases between 24 and 72 hours except when 8.7 ppb Ag and 791.7 ppb Zn were released from the 67% Ag/33% Zn CWF_{DISKS}. With that said, less regrowth was found as eluted Ag and Zn concentrations increased, as shown by the darkest contours remaining in the upper right corner of Figure 5.7C. Similarly, within the batch phase, regrowth effects were most acute when Ag was in concentrations less than 10 ppb (and particularly when less than 5 ppb) and Zn was in concentrations less than 200 ppb, as evidenced by the expansion of the lightest contours in the bottom left corners of Figure 5.7D-F. Yet, like was found among filters, higher Ag and Zn concentrations led to improved or maintained disinfection over time. More specifically, within batch experiments, 20 ppb Ag and 160 ppb Zn achieved LRVs (Log C_o/C) of 2.1 ± 0.9, 2.4 ± 1.0, and 2.5 ± 0.9 (i.e., disinfection) after 24, 48, and 72 hours, respectively, while pot-equivalent elution values of 20.5 ppb Ag and 377 ppb Zn achieved an LRV of 1.9 ± 0.1 at the same time points, respectively. An overarching consistency is thus observed across experiments, highlighting some translation between experimental phases. Further, these results demonstrate that including Zn within CWFs for enhanced disinfection may present significant opportunity to reduce filter cost while maintaining bactericidal efficacy. The translation between phases is, however, clearly imperfect. The trends found during batch phases are demonstrably more direct and linear, which illustrates that additional factors are contributing to bactericide within filters.

This inconsistency between batch and filtration experiments is moreover particularly clear in Figure 5.8, which shows the relationship between LRV (Log C_o/C) with the product of

metal concentration and time (i.e., CT) within the first 24 hours of testing. Specifically, Figures 5.8A, 5.8B, and 5.8C investigate the impact of total metal concentrations (i.e., ppb Ag + ppb Zn) at 10^2 CFU/mL, 10^3 CFU/mL, and without aggregation by initial bacterial concentration, respectively. Figure 5.8D further illustrates the CT-LRV relationship when only Ag is present, and Figure 5.8E illustrates the same relationship found by the CWFs in storage (as illustrated in Figures 5.7A-C). The results highlight that batch experiments broadly followed the Chick-Watson disinfection model regardless of bacterial concentration, though better alignment was observed at the lower bacterial concentration and when only Ag was present. However, the CWF regression achieved significantly less predictive capacity, likely resulting from two factors. First, differences in NP forms between batch and filter phases may have led to different disinfection efficacies. That is, batch-phase experiments used direct nanoparticle injection, suggesting ion penetration, nanoparticle-cell interactions, and oxidative killing all contributed to bacterial deactivation (Venis and Basu 2020). Meanwhile, the ICP-MS acidification procedure produces inherent uncertainty regarding which forms of silver and zinc were present within the CWF effluents. Which mechanisms were responsible for bactericide, as well as their associated efficacies, thus remains unknown. Further investigation of eluted metal forms in co-fired CWF effluents is required. Second, it is also likely that non-Ag-Zn elements inhibited or enhanced disinfection within the CWF effluents. For instance, previous research has highlighted that several transition metals, some of which are common within clay materials (e.g., Al_2O, MgO), can interact with Ag to improve or inhibit disinfection (Garza-Cervantes, et al. 2017, Venis and Basu 2020). Future investigation of the complete composition of CWF effluent water matrices,

including Zn, Ag, and other elements unenumerated in this work, is moreover recommended. Better characterization of background elemental compositions within various clays may provide necessary insights for elucidating CWF disinfection mechanisms during storage.

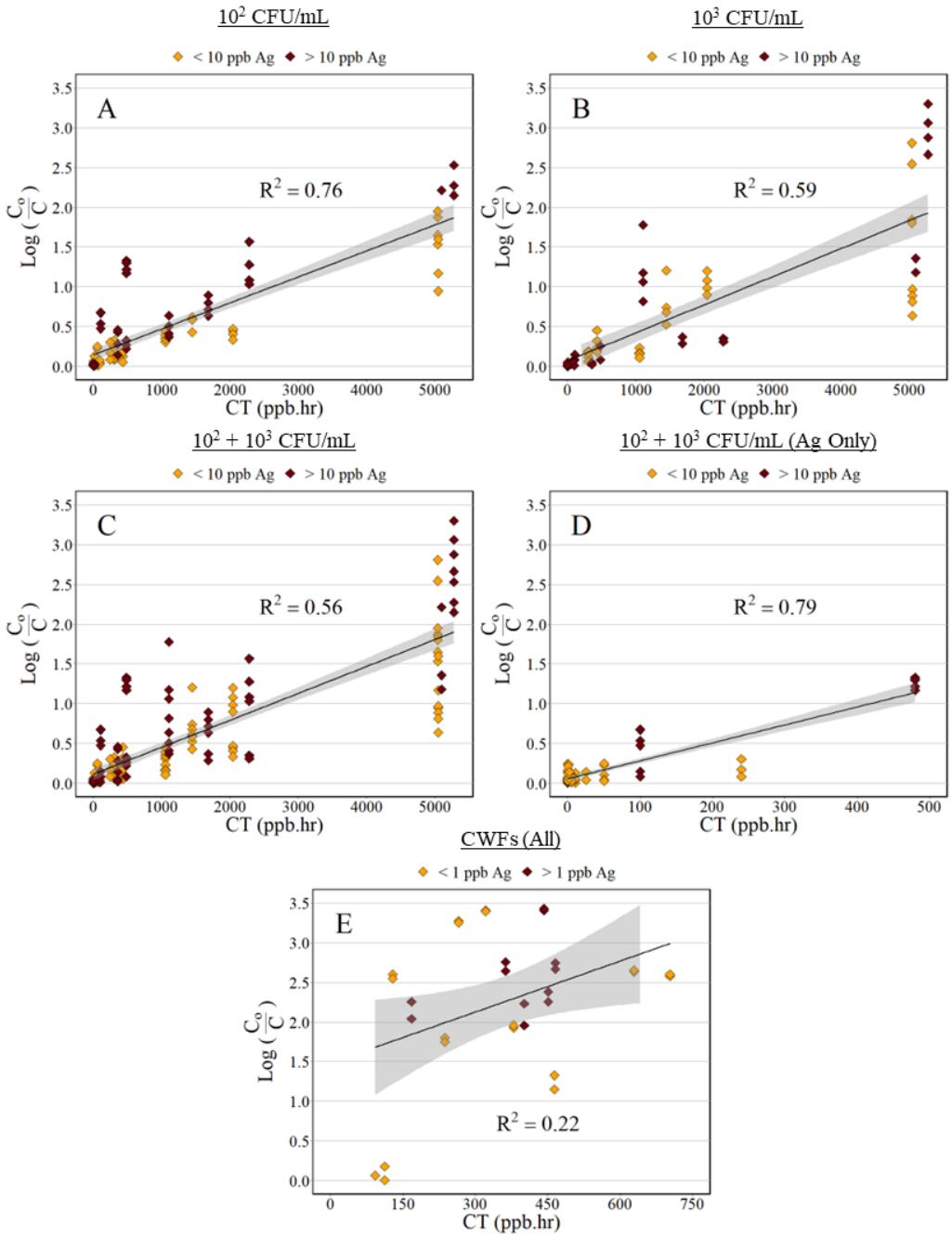

Figure 5.8. Log Removal Value (LRV) versus the product of nanoparticle concentration and time (CT) within 24 hours when Ag and Zn are combined in batch-phase tests with (A) 10^2 CFU/mL, (B) 10^3 CFU/mL, and (C) when disaggregated by bacterial concentration, as well as when (D) only Ag is present (disaggregated bacterial concentration). Results from CWFs (E) during storage after filtration also presented. Note: shaded region is 95% confidence interval on regression line.

The combination of inconsistent metal releases from filters, inconsistent translations between metal impregnation and elution, and associated changing bacterial concentrations during storage furthermore highlights that uncertainty exists regarding water quality during long-term storage. In practice, these results suggest CWF manufacturers must ensure size exclusion processes remain robust such that the bacteria passing through the matrix is limited and subsequent risk of regrowth is minimized. If not managed appropriately, usage protocols may require waiting for water quality to improve in storage after filtration, yet not so long as to potentially observe bacterial regrowth, potentially creating additional time and effort burdens that could facilitate misuse or disuse (Ray and Smith 2021, Burt, et al. 2017). More research in this regard is suggested.

5.4 Conclusions

The objective of this study was to demonstrate how metallic impregnation translates to associated CWF disinfection during storage, as well as highlight the efficacy of AgNP with ZnO supplementation. The research also aims to illustrate the respective roles of filtration and impregnated metal disinfection in CWF water treatment to provide insight into appropriate usage. During CWF experiments, bacterial water safety reached approximately 2 LRV (removal) post-filtration, which improved significantly after 24 hours of storage. These improvements were further largest among filters impregnated with either Ag, Zn, or both, suggesting eluted MNPs do indeed maintain water safety during storage.

Batch experiments showed that Zn inclusion enhanced Ag disinfection efficacy, emphasizing its potential value for CWF optimization. For instance, in water with 10^2 CFU/mL, 1 ppb Ag

and 800 ppb Zn achieved an LRV 2.0 (disinfection) after 24 hours, while 10 ppb Ag and 160 ppb Zn achieved an LRV of 1.75. In other words, a 5-fold increase in Zn allowed for 10-fold reduction in Ag while maintaining LRV, demonstrating clear synergistic disinfection. Required Ag elution concentrations can thus be reduced significantly if sufficient Zn concentrations are present and effluent bacterial concentrations are controlled. How to ensure such Zn concentrations, however, requires further investigation. That is, Ag impregnation translated well to measured elution, and further accurately scaled from CWF_{DISKS} to the CWF_{POT}. Meanwhile, eluted Zn concentrations were not related to impregnation and differed significantly between disks and the pot. These observations indicate background Zn presence, suggesting clay elemental composition is more relevant to CWF disinfection during storage than previously considered.

Key outcomes include:

- Zn inclusion in CWFs simultaneously maintained or improve disinfection while reducing cost
- MNP contribution to disinfection was greatest between 24-48 hours of storage, with reductions in water quality by 72 hours. Consumption of produced water within 48 hours represents an idealized storage duration.
- Additional research is recommended to characterize CWF effluent matrices and identify an array of eluted metals that may contribute to disinfection.

5.5 Acknowledgements

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Chapter 6: Ceramic Water Filter Material Design Optimization

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Abstract:

Ceramic water filters (CWFs) are decentralized water treatment technologies often distributed in rural and resource-restricted contexts. Breakage and slow flowrates, however, commonly lead to disuse. Increasing filtrate production and material strength could thus improve CWF longevity. This research aims to identify key design parameters of such an optimal filter. A central composite design is used to evaluate CWF disks with 38-50% clay (by volume) and nominal sawdust particle sizes ranging between 300-420 μm. Nested stepwise multiple regression analysis is then used to estimate flow, bacterial removal, and flexural strength with generalized parameters to facilitate model scalability and potentially improve quality control processes. Possibility-based design optimization (PBDO) resultingly predicts filter characteristics that maximize flowrate while maintaining acceptable disinfection and strength. Nested modeling with porosity, intrinsic permeability, and dry density proved valid, yielding R^2 values of 0.97, 0.76, and 0.68 for flowrate, bacteria

removal, and strength, respectively. Meanwhile, models which used clay proportion and sawdust size as direct inputs were heteroscedastic and invalid. Using these generalized parameters to predict CWF performance may therefore offer opportunity to improve quality control procedures at low cost. PBDO estimates further suggest CWF pot flowrates can reach above 8 L/hr while meeting LRV and strength requirements if porosity is less than 48%, density is more than 1.2 g/cm^3, and intrinsic permeability is less than 29 x 10^{-9} cm^2. This may be achieved by increasing sawdust proportion and decreasing sawdust size from the model center point, which is an existing CWF design.

Keywords:

household water treatment, material optimization, point of use water treatment, quality control, machine learning

Highlights:

- Ceramic filter flowrate can increase without compromising strength or disinfection
- Porosity, dry density, and intrinsic permeability are useful predictors of filter performance
- Fuzzy logic is valuable for predicting filter performance in presence of uncertainty
- Optimal filters should have 40% < %clay < 47% and sawdust sized < 600 μm

6.1 Introduction

Ceramic water filters (CWFs) are decentralized, point-of-use (POU) drinking water treatment technologies widely distributed and used in resource-restricted contexts (Venis and Basu 2020). They are manufactured in more than 50 factories worldwide (Rayner, et al. 2017) and are regarded by diverse water, sanitation, and hygiene (WaSH) experts as a useful tool for addressing challenges with access to safe drinking water in the immediate term (CMWG 2010, WHO; UNICEF 2017). Specifically, CWFs are promoted due to their simple usage and maintenance protocols, lower cost relative to other POUs and centralized water treatment solutions, accessible manufacturing materials, and general compliance with international drinking water guidelines for bacteria removal from household water filters (Sobsey, et al. 2008, Venis and Basu 2020, WHO 2011). However, certain design features have been shown to cause disuse in field settings and limit proliferation. For instance, both Burt et al. (2017) and Salvinelli et al. (2017) found 40% of study participants stopped using their CWF because the flowrate was too slow to meet their household needs. These observations further align with expectation as CWFs typically produce 1-4 L/hr, while the World Health Organization (WHO) stipulates that approximately 40 L of safe water is needed per household (5 people) per day for only "survival, basic hygiene, and basic cooking" (Reed and Reed 2013). Additionally, Brown et al. (2009), Roberts (2004), and Lemons et al. (2016) found 60%, 25%, and 18% of distributed filters broke within 48 months, 11 months, and 6 weeks, respectively. Filter breakage was further the leading cause of disuse in all studies. Flowrate and material strength are demonstrably important determinants of CWF longevity and acceptability as a drinking water solution. Improving these performance

metrics is thus critical to facilitating increased CWF adoption and consequently advancing progress towards universal access to safely managed drinking water services – Sustainable Development Goal (SDG) 6.1.

CWFs are fabricated by combining clay and a firing material (FM; e.g., sawdust, flour, rice husks) into a dry homogenous mixture, which is subsequently combined with water to create a plastic and pliable "dough". The dough is then pressed in a mold to form the desired shape (most typically a pot) and dried for between 1-6 weeks depending on the geography and season. Once completely dry, filters are placed in a kiln and fired up to 900-1000 ºC over 12-18 hours until the FM is burned away and the clay is vitrified (CMWG 2010). The voids created by the burnt-out material resultingly form a porous network throughout the filter body that physically removes contaminants by size exclusion. The ratio between clay and FM, as well as the FM particle size, are thus related to flowrate, bacteria removal, and strength.

However, studies that have evaluated the relationships between clay-FM proportions and FM size on the abovementioned performance metrics have reported a wide range of somewhat inconsistent results. For instance, Rayner et al (2017) found disk-shaped filters (Ø = 10 cm) with 13.7% sawdust by weight (~40% by volume) and a nominal sawdust particle size of 1.78 mm achieved an average flowrate of 70 mL/hr and average LRV of 1.9. Those with the same sawdust proportion and nominal sizes of 0.595 mm and 0.425 mm further achieved average flowrates of 27 mL/hr and 17 mL/hr and average LRVs of 4.4 and 2.1, respectively. When sawdust proportion was increased to 17% by weight (~47% by volume) but nominal sizes were kept at 0.595 mm and 0.425 mm, average flowrates were 100 mL/hr and 17 mL/hr

and average LRVs were 2.4 and 4.0, respectively. Similarly, van Halem et al. (2017) showed filter pots with 24% and 31% rice husks by weight (nominal size = 0.5 mm) achieved average flowrates of 3.0 L/hr and 19.0 L/hr, respectively, while both achieving average LRVs of 0.9. Soppe et al. (2015) also found pot-shaped filters with 31% rice husk by weight and nominal particle sizes of 0.5 mm and 0.75 mm achieved average flowrates of 3.0 L/hr and 10.1 L/hr, average LRVs of 2.8 and 0.8, and average moduli of rupture (MOR; strength measure) of 2.4 MPa and 1.4 MPa, respectively. Pots with 39% risk husks sized at 0.5 mm achieved an average flowrate of 12 L/h, LRV of 4.1, and MOR of 1.6 MPa, respectively. Finally, Servi et al. (2013) found that small CWF disks (Ø = 2 cm) with 20% rice husks by weight and nominal sizes of 0.39 mm, 0.50 mm, and 0.65 mm achieved average flowrates of 14 mL/hr, 13 mL/hr, and 69 mL/hr, LRVs of 3.0, 2.0, and 2.8, and loads at rupture of 0.9 kN, 1.1 kN, and 0.5 kN, respectively. Evaluation of these results together furthermore illustrates a general trend of increasing flowrate with corresponding increases in FM proportion and size, while no clear trend is observed for LRV. Flowrate and LRV are also notably unrelated. Likewise, strength appeared to generally decrease with increasing FM sizes and proportions, though this trend was also not always consistent from one evaluated level to the next. Identifying an optimal CWF design from this data which may meet user drinking water quantity needs while also achieving acceptable disinfection and robust material strength is thus demonstrably challenging. With that said, very few studies have assessed filter strength, making broad inferences on this relationship difficult to ascertain. Additional research into how these CWF input parameters relate to strength is required.

The observed unclarity with respect to how LRV changes with changes to input parameters is further another matter requiring additional exploration. LRV is evidently more impacted by factors beyond the size and amount of FM within a CWF mixture than flowrate, as size exclusion processes depend not only on the available space through which water can pass but the size and tortuosity of that space relative to the size, shape, and flow path of a contaminant (Venis and Basu 2020). Meanwhile, site-specific factors such as physicochemical clay properties, firing methods and kiln organization, atmospheric/climatic conditions, and mixing technique can all impact how FM burns away, how much burns away, and how pore sizes consequently form and connect (CMWG 2010, Soppe, et al. 2015). As such, the filter production process is inherently chaotic and small differences between each individual filter's initial conditions can lead to unique material properties. Large variances in disinfection among filters with the same FM sizes and proportions, both within and between factories, may resultingly ensue. The significance of this randomness is moreover most acute within quality control (QC) and quality assurance (QA) practices. Namely, many factories rely on flowrate, FM size, and FM proportion for predicting LRV despite this approach's proven inaccuracy (Rayner, et al. 2017, van Halem, van der Laan and Soppe, et al. 2017). An alternative framework for modeling disinfection with generalized filter characteristics that account for these uncontrolled factors may therefore offer opportunity to more accurately predict filter performance and identify an optimum design that can be applied across manufacturing facilities.

This study furthermore applies theory from computational fluid dynamics (CFD) research on convection in porous media to model flowrate (Q), LRV, and MOR in CWFs with

generalized material parameters. Specifically, model parameterization uses established generalized characteristics for estimating flow through porous media (Nield and Bejan 2013) which may also be easily and/or inexpensively measured. These models are subsequently utilized to identify optimum (or a range of optimum) filter characteristics that maximize flowrate while maintaining adequate disinfection and strength.

6.2 Methodology

All CWFs used in this research were produced by Safe Water Ceramic of East Africa in Arusha, Tanzania in June 2019 and transported to Carleton University in Ottawa, ON, Canada for testing in July 2019.

6.2.1 Disk Filter Manufacturing Procedure

CWFs were made with raw clay mined near Babati, Tanzania, and locally sourced pinewood sawdust (firing material). Sawdust was first passed through either a No. 20 (841 μm mesh) or No. 30 (595 μm mesh) sieve (Figure 6.1A) and subsequently combined in appropriate proportions, as per Table 6.1. Note particles that passed through No. 20 and No. 30 meshes are hereafter referenced as big sawdust (BSD) or small sawdust (SSD) particles, respectively. Clay and sawdust mixtures were then added volumetrically to a plastic-lined container and mixed by hand with a ball whisk for a minimum of 15 minutes until visually homogeneous (Figure 6.1B). Water was added incrementally until the desired dough consistency was achieved, as per the local potters' expertise and judgements (approx. 20% of dry mixture volume). The wet dough was kneaded by hand (Figure 6.1C) for additional mixing, after which a portion was added to a rectangular mold and consolidated with a plastering trowel

to reach a thickness of 2.5 cm. Four disk filters were then cut out of the mold with a 7.5 cm diameter piece of PVC pipe (Figure 6.1D), labelled according to the names found in Table 6.1, and air-dried for four weeks (Figure 6.1E). Once deemed sufficiently dry by the potters, all 60 disks were added to a packed kiln (Figure 6.1F; disks are scattered in between the pots) and fired to approx. 960 ºC over 12 hours. After firing, filters were shaved with a coarse file to fit within the testing unit, reaching an approximate diameter and thickness of 7 cm and 2 cm, respectively. Filter top and bottom surfaces were measured with a bubble level to ensure equal and consistent disk thicknesses were achieved.

Figure 6.1 CWF Disk Preparation Process

Table 6.1 CWF disk material inputs and CCD coordinates

Name	%Clay	%Sawdust	%Big Sawdust	%Small Sawdust	$^1\zeta_1$	ζ_2	$^2x_1$	x_2
$C_{38}B_{100}$	38	62	100	0	38	100	-1.0	1.0
$C_{38}B_{60}$			60	40	38	60	-1.0	0.2
$C_{38}B_{50}$			50	50	38	50	-1.0	0.0
$C_{38}B_{40}$			40	60	38	40	-1.0	-0.2
$C_{38}B_0$			0	100	38	0	-1.0	-1.0
$C_{44}B_{100}$	44	56	100	0	44	100	0.0	1.0
$C_{44}B_{60}$			60	40	44	60	0.0	0.2
$C_{44}B_{50}$			50	50	44	50	0.0	0.0
$C_{44}B_{40}$			40	60	44	40	0.0	-0.2
$C_{44}B_0$			0	100	44	0	0.0	-1.0
$C_{50}B_{100}$	50	50	100	0	50	100	1.0	1.0
$C_{50}B_{60}$			60	40	50	60	1.0	0.2
$C_{50}B_{50}$			50	50	50	50	1.0	0.0
$C_{50}B_{40}$			40	60	50	40	1.0	-0.2
$C_{50}B_0$			0	100	50	0	1.0	-1.0

$^1\zeta$ = natural variable; ^2x = coded variable

6.2.2 Material Characterization

6.2.2.1 Intrinsic Permeability, κ

Each filter's intrinsic permeability was evaluated according to a temperature-normalized Darcy's Law (Equation 6.1).

$$\kappa = \frac{v\mu_{w,^oC}}{g\rho_{w,^oC}\Delta P} \qquad [6.1]$$

Where κ is intrinsic permeability (cm^2), v is the hydraulic velocity ($cm\ s^{-1}$), $\mu_{w,^oC}$ is the dynamic viscosity of the water at the temperature of testing (Pa s), g is the gravitational force ($cm\ s^{-2}$), $\rho_{w,^oC}$ is the density of water at the temperature of testing ($g\ cm^{-3}$), and ΔP is the pressure gradient from the top to bottom surface of the filter (cm).

For measurement, each filter disk was attached to the testing cylinder following the experimental preparation procedure described in Section 6.2.3.1 using distilled water. The hydraulic velocity was calculated as $v = V_w/A_D$ where V_w is the volume of water that passed through the filter in 1 hour (cm^3hr^{-1}), and A_D is the disk area (cm^2). κ measurements for each disk filter were repeated according to the disk's position within the CCD, as per Figure 6.2.

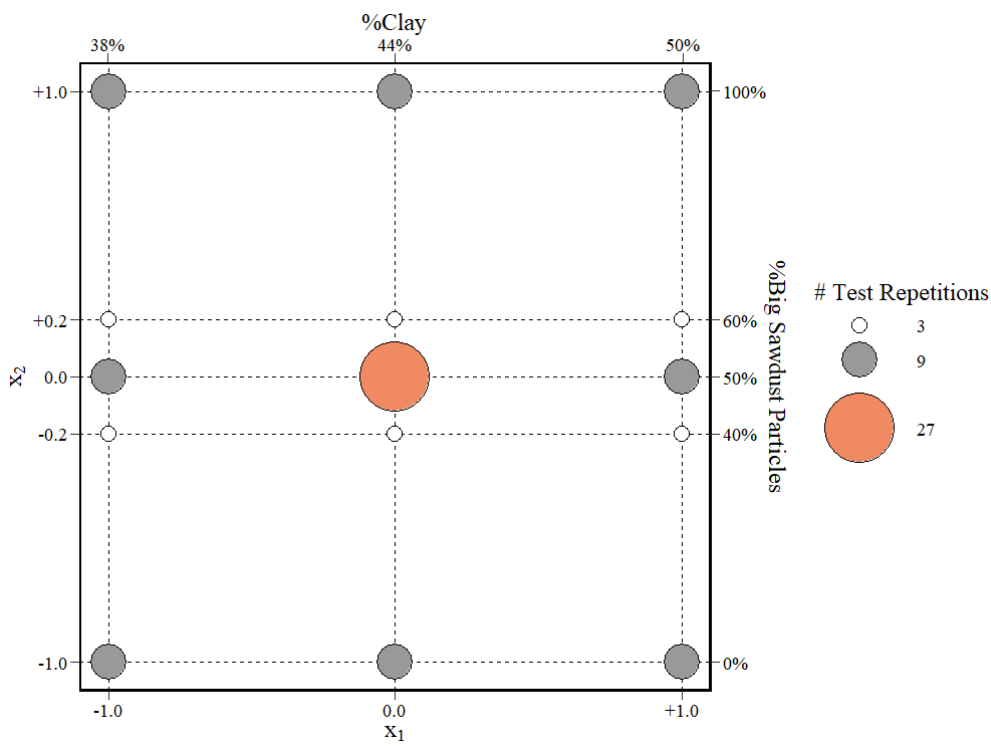


Figure 6.2 Face-centred central composite design structure

6.2.2.2 Porosity, φ

Filter porosity was determined using open-source ImageJ software, as shown in Figure 6.3. Specifically, once all strength testing had been completed (see Section 6.2.3.2), inner surfaces were imaged using a Nikon Digital Single-Lens Reflex (DSLR) camera (Figure 6.3A). A section of the image was then cropped and edited for exposure, contrast, and clarity

in MS Photos (Figure 6.3B). Each disk was photographed a minimum of 6 times with focused attention on different sections to capture as much pore detail as possible. Different sections of the disk were also cropped for each porosity estimate. All images were subsequently uploaded into ImageJ and further processed to remove any shadows and unclear areas (Figure 6.3C). Once complete, the Threshold function measured porosity using the images' greyscale spectrum; the filter porosity was measured as the peak of this spectrum (Figure 6.3D). This process was then repeated between 2-18 times per disk (6-54 times per type) depending on the filter position within the CCD design.

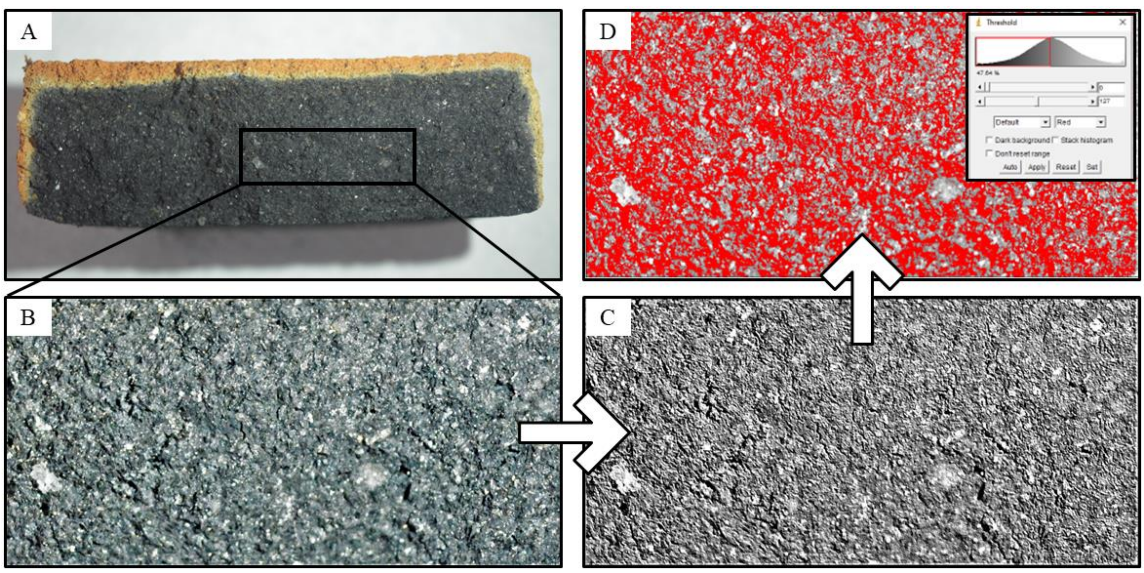


Figure 6.3 Porosity Measurement Procedure

6.2.2.3 Density, ρ_D

Before any testing began, all filters were placed in an incubator at 65 °C for at least 48 hours to release any residual moisture within the matrix. Filter mass was then measured for each filter disk after gently brushing the surfaces to remove any solids which may have

accumulated during transportation and/or storage. Density was further calculated by dividing the measured mass by the filter disk volume, as per Equation 6.2.

$$\rho_D = \frac{M_D}{V_D} \quad [6.2]$$

6.2.2.4 Young's Modulus, E

Young's modulus was calculated as the slope of the elastic region on the stress-strain curve for each filter. Stress was calculated as the applied load (N) at time t divided by the disk filter area (mm^2), while axial strain was calculated as the amount of compression measured by the Instron 5582 (mm) relative to the initial measured disk thickness (mm).

6.2.3 Testing Procedure

6.2.3.1 Flow/Bacterial Testing

A full description of disk filter preparation and testing methods may be found in Chapter 5. Briefly, each filter disk was first brushed with a hard bristle and washed with membrane-filtered distilled water to remove any surface grime. The disk was then added to a 1L beaker of boiled distilled water for 30-60 minutes, after which it was placed in a in an incubator at 65 ^{o}C for a minimum of 48 hours. Once dried, the disk's outer edges were wrapped tightly with Teflon tape to encourage one-directional flow and sealed in a threaded holding unit with silicon glue at the top and bottom edges. After a minimum of 12 hours of curing, the holding unit containing a disk was screwed into the testing cylinder (PVC piping) until watertight. The testing cylinder was subsequently filled with 25 cm of constant head. A timer was started

after at least 30 mL (approx. 0.5-1 pore volume) passed through the filter and disks were left to flow for 60 minutes uninterrupted. Filtrate was measured by mass with a precision balance.

6.2.3.1.1 Bacterial Growth and Monitoring

Non-pathogenic K-12 *Escherichia coli* (ATCC® 29947™; Migula; Catellani and Chalmers) was used as the indicator species for bacterial removal efficiency. 1 μL of frozen stock solution (see Chapter 4) was added isometrically to 9900 μL of No. 3 Nutrient Broth (i.e., 0.1% solution) and incubated at 37 ºC for 19 hours, approximately halfway through the stationary growth phase. An approx. 10^8 CFU/mL final concentration was resultingly achieved.

Before each test, cultured solution was added volumetrically to synthetic soft water, as per Table 7 in USEPA (2002), to reach a concentration between 10^5 and 10^6 CFU/mL. All measured water quality characteristics may be found in Table 6.2. A high initial bacterial concentration was chosen to reduce the likelihood of a limited maximum LRV. Influent bacterial concentrations were sampled from the testing cylinder immediately after the timer was started and effluent concentrations were sampled from the collection vessel after 60 minutes of filtration. Bacterial concentration was enumerated using membrane filtration as per USEPA Method 1603/4 with mTEC nutrient agar (BD Microbiology). All influent and effluent samples were taken in duplicate.

Table 6.2. Influent Water Quality

Water Quality Parameter	Value (±SD)
pH	7.38 ± 0.03
ORP (mV)*	-21 ± 3
Dissolved Oxygen (mg/L)	8.62 ± 0.16
Temperature (^{O}C)	22.2 ± 0.8
Alkalinity (mg/L $CaCO_3$)	29 ± 2
Hardness (mg/L $CaCO_3$)	23 ± 2
*TOC** (mg/L)*	6.1 ± 0.2
E. coli (x10^5 CFU/mL)	2.38 ± 0.88

*ORP = Oxidation Reduction Potential
**TOC = Total Organic Carbon

6.2.3.2 Strength Testing

Filter strength was evaluated using a modified ASTM C1499 – 19. Concentric ring loading units were machined at Carleton University's Civil Engineering workshop to the specified dimensions shown in Figure 6.4. Before testing, upper (Figure 6.4A) and lower (Figure 6.4B) load supports were attached to the respective holding units of an Instron 5582 compressive testing apparatus. Disks were centred atop the lower load support and checked to ensure they were level, after which the upper load support was lowered to just above the filter surface. Disks were then loaded at a rate of 10 mm/s until fracture.

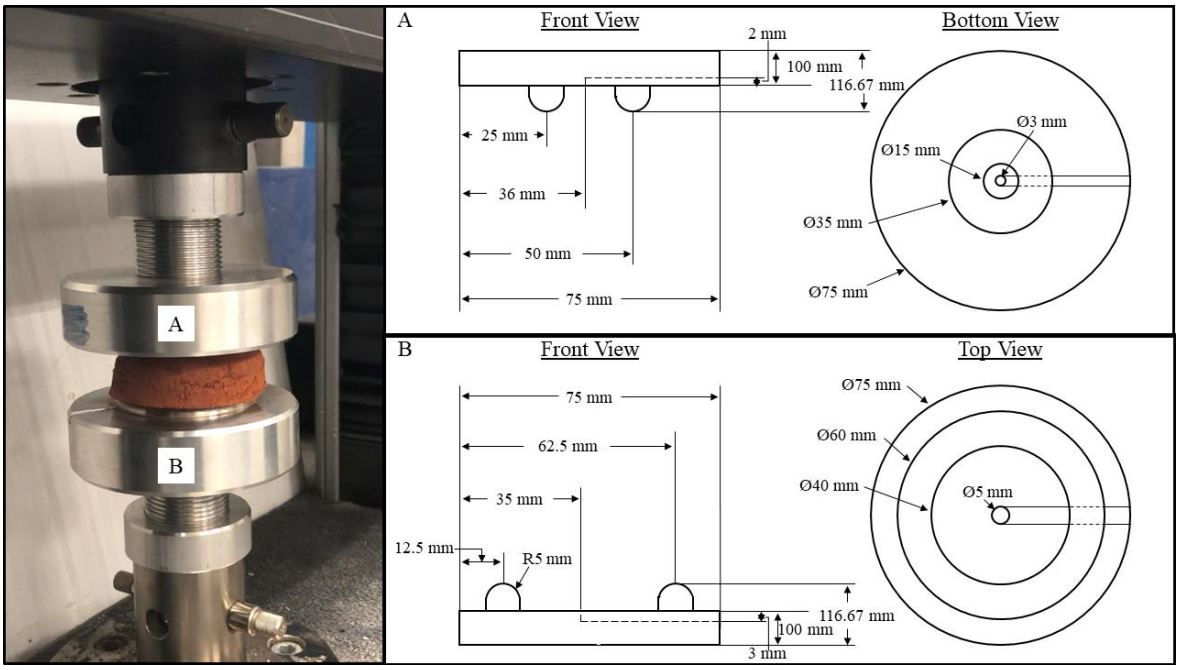


Figure 6.4 Strength Testing Apparatus

As per the described method, the filter modulus of rupture (MOR) was calculated using Equation 6.3. Note that this calculation differs from common MOR calculations in that it is divided by 2π due to the circular nature of the concentric rings. This approach was chosen to account for commonly stated limitations with conventional 3-point flexural testing procedures including non-uniform stress distribution in cylindrical ceramic samples and overestimated strengths (Morell 1998, Lupercio, et al. 2021).

$$MOR = \frac{3P}{2\pi h^2} \qquad [6.3]$$

P (N) is the breaking load in units of N and h (mm) is the specimen thickness.

6.2.3.3 Experimental Structure

The flowrate and bacterial removal experiments were examined using a face-centred central composite design (FC-CCD). Due to the nonrepeatable (i.e., destructive) nature of strength testing, each of the sixty disks were ruptured once only. Figure 6.2 illustrates the FC-CCD structure with reference to the number of repetitions taken for each disk type based on their clay-sawdust (C:SD) ratios and BSD:SSD ratios, as well as their coded positions as referenced in Table 6.1. As per CCD experimental convention, the centroid was evaluated three times more than the corners, which were evaluated three times more than interim nodes (Montgomery 2009). The centroid used in this study is the existing filter design used by the manufacturing partner.

6.2.4 Design Optimization

6.2.4.1 Model Determination

Models for both input parameters and performance metrics were determined using back-facing stepwise multiple regression analysis. Parameter inclusion criterion was set as $p<0.05$, while the exclusion criterion was $p>0.10$. Each input could be raised to a maximum of the 4th degree and all interaction terms were included. Models were also evaluated with direct, log, or squared responses. The model with the lowest Root Mean Square Error (RMSE) and lowest Standard Error (SE) was then chosen, so long as the p-value of the Shapiro-Wilk Statistic for residual normality was >0.10. Filter characteristics κ, φ, and ρ_D were modeled in terms of the percentage clay included (C) and percentage of big sawdust particles included (BSD), while LRV, MOR, and Q were modeled in terms of κ, φ, and ρ_D. This nested structure facilitates model scalability such that performance outputs may be evaluated in terms of

general characteristics of porous media rather than specific manufacturing inputs. Additionally, LRV, MOR, and Q were also modeled using the same methodology in terms of C and BSD for comparative purposes. The normality constraint was not, however, included for these latter models due to observed heteroscedasticity (see Section 6.3.3.1).

Please also note that Young's Modulus, E, was measured but not modeled due to the specialized equipment required. That is, measuring E was considered beyond the technological and financial capacities of most CWF manufacturers and its inclusion would therefore limit the models' potential universality and scalability across diverse facilities.

6.2.4.2 Possibility-Based Design Optimization

PBDO has recently emerged as a valuable alternative to Reliability-Based Design Optimization (RBDO) for stochastic engineering design when data limitations exist. Namely, RBDO employs a performance measure approach (PMA), which determines the feasibility for each constraint while assuming a specified limit failure probability. Specifically, PMA calculates the minimum distance between a candidate design and a constraint boundary and then uses the candidate's probability-distribution function (PDF) to identify a new value at which the failure probability is satisfied (Choi, et al. 2006). RBDO then identifies new constraints from those probabilities. The method therefore necessarily requires sufficient data to create accurate and precise PDFs to characterize uncertain design variables, which was neither feasible nor possible in the present work. As such, PBDO instead creates uncertainty intervals around deterministic model predictions using fuzzy logic. PBDO thus follows a modified PMA, where rather than check the feasibility using a PDF, the feasibility

is checked according to where the optimum lies within the range of possibilities relative to the ranges of possibilities for constraining parameters. This notion may be expressed mathematically as

$$minimize\ size(\boldsymbol{d})$$

$$subject\ to\ G_{\Pi i}[\boldsymbol{d}(\boldsymbol{X})] \leq C_i; \quad i = 1, 2, \ldots, np$$

$$\boldsymbol{d^L} \leq \boldsymbol{d} \leq \boldsymbol{d^U}$$

where **d** is the design vector, defined as $\boldsymbol{d} = [d_i^T] \in R^n$; **X** is the vector of fuzzy variables, defined as $\boldsymbol{X} = [X_i]^T \in R^{nr}$; $G_{\Pi i}$ is the *i*th possibility constraint; C_i is the constraint bound; $\mathbf{d}^L$ and $\mathbf{d}^U$ are the lower and upper constraint bounds of the design vector, respectively; and *n, nr,* and *np* are the number of design variables, fuzzy variables, and possibility constraints, respectively. Note too that superscript T refers to a transposed term. Plainly, this function describes the systematic minimization of an identified design variable of interest given that the constraints remain below some threshold, C_i. The model therefore checks all positions within the fuzzy intervals to ensure the acceptable criteria are always met, even in a *worst-case* scenario. These checks are further completed by iteratively multiplying the deterministic outputs by parameter-specific correction factors within the possibility bounds until the constraint criteria are violated.

In the present study, the optimized values were found using a sequential optimization method. The deterministic optimum was found at the maximal constraints and the uncertain parameters were varied thereafter to find new constraints (i.e., the correction factor at which at least one constraint is violated) using the PMA. This process was then repeated until the

solution converged towards a single outcome under which all conditions were satisfied. The optimization method further used Sequential Least Squares Programming (SLSQP) in Python with the scipy.optimize package based on Kraft (1998). Flowrate, Q, was chosen as the design variable with the objective of its maximization (i.e., minimization of -Q). LRV was constrained to be always > 2.0, as per WHO guidelines for household-scale filtration technologies (WHO 2011), and MOR was constrained to always be > 1.2, the mean value achieved by the centroid, the manufacturer's current design. Fuzzy intervals were determined from the model standard errors (as %) for each parameter. Deterministic, maximal flow, and safest-case predictions are reported.

6.3 Results and Discussion

6.3.1 Material Characterization

Shown in Table 6.3 are the measured material parameters for each CWF disk type. Both clay and BSD proportions influenced material characteristics, though with differing effects. For instance, a near-linear change in average porosity was observed with clay proportion between filters with the same BSD proportions; $C_{38}B_{100}$ was measured as 26.5% larger than $C_{44}B_{100}$, which was measured as 29.4% larger than $C_{50}B_{100}$. Meanwhile, the $C_{38}B_{100}$ filters had a measured average κ 1411% larger than that of $C_{44}B_{100}$, which had a 118.6% larger average κ than $C_{50}B_{100}$ filters. Similarly, filters with 38% clay had a Young's Modulus 124% lower than those with 44% clay, which had a Young's Modulus only 40% lower than those with 50% clay. As such, while the amount of pore space within each filter changed relatively consistently across all three levels, the same change in FM proportion led to an increase in

the fluid penetrability by more than a factor and a n 85% decrease in material stiffness. This observation further aligns with outcomes measured by Rayner et al (2017), who found no correlation between porosity and permeability, but did find that the C:SD ratio significantly impacted both. When considering the differences in E, these results also highlight that an inflection point may exist between 38% and 44% clay where permeability may be increased while material strength remains adequate.

Table 6.3. Disk Filter Material Characteristics (C is % clay and B is % large sawdust particles)

Name	Nominal Pore Diameter (μm)	Diameter, Ø (cm)	Thickness, t (cm)	Porosity, φ (%)	Dry Density, ρ_D (g/cm^3)	Intrinsic Permeability, κ (10^{-8} cm^2)	Young's Modulus, E (MPa)
$C_{38}B_{100}$	420	7.1 ± 0.05	2.0 ± 0.11	58.2 ± 0.00	1.0 ± 0.03	14.2 ± 15.5	0.37 ± 0.09
$C_{38}B_{60}$	400	7.1 ± 0.05	2.2 ± 0.17	55.2 ± 2.38	1.0 ± 0.05	7.63 ± 8.12	0.52 ± 0.23
$C_{38}B_{50}$	360	7.1 ± 0.10	2.1 ± 0.08	56.1 ± 3.76	0.9 ± 0.04	15.0 ± 15.7	0.34 ± 0.08
$C_{38}B_{40}$	345	7.1 ± 0.00	2.1 ± 0.17	59.8 ± 1.20	0.9 ± 0.03	17.0 ± 17.9	0.34 ± 0.20
$C_{38}B_{0}$	300	7.2 ± 0.09	2.2 ± 0.05	53.5 ± 1.52	1.1 ± 0.05	6.79 ± 6.95	0.64 ± 0.25
$C_{44}B_{100}$	420	7.2 ± 0.00	2.0 ± 0.11	46.0 ± 1.07	1.2 ± 0.02	0.94 ± 0.97	0.91 ± 0.27
$C_{44}B_{60}$	400	7.2 ± 0.05	1.9 ± 0.25	45.0 ± 2.18	1.2 ± 0.05	0.50 ± 0.54	0.82 ± 0.08
$C_{44}B_{50}$	360	7.1 ± 0.05	2.1 ± 0.10	43.2 ± 1.26	1.3 ± 0.04	0.23 ± 0.25	1.17 ± 0.59
$C_{44}B_{40}$	345	7.2 ± 0.00	2.2 ± 0.10	44.6 ± 2.41	1.3 ± 0.05	0.32 ± 0.33	1.14 ± 0.22
$C_{44}B_{0}$	300	7.2 ± 0.00	2.0 ± 0.06	44.1 ± 1.45	1.3 ± 0.04	0.36 ± 0.41	0.96 ± 0.16
$C_{50}B_{100}$	420	7.2 ± 0.09	2.1 ± 0.12	35.4 ± 1.24	1.2 ± 0.03	0.43 ± 0.45	1.42 ± 0.29
$C_{50}B_{60}$	400	7.2 ± 0.05	2.2 ± 0.00	38.4 ± 2.35	1.2 ± 0.02	0.24 ± 0.26	1.39 ± 0.37
$C_{50}B_{50}$	360	7.1 ± 0.08	2.2 ± 0.18	36.1 ± 2.55	1.3 ± 0.05	0.23 ± 0.24	1.19 ± 0.40
$C_{50}B_{40}$	345	7.2 ± 0.08	2.2 ± 0.10	38.1 ± 2.72	1.2 ± 0.05	0.30 ± 0.33	1.53 ± 0.71
$C_{50}B_{0}$	300	7.1 ± 0.07	2.1 ± 0.10	37.4 ± 0.74	1.3 ± 0.03	0.28 ± 0.30	1.47 ± 0.50

The impacts of BSD proportion on measured characteristics were also broadly nonlinear and ambiguous. For example, both ρ_D and φ increased with increasing BSD proportions, though these increases were only significantly different from each other among filters with 38% clay ($p<0.01$) and 44% clay ($p<0.01$). BSD proportion did not impact density or porosity among filters with 50% clay ($p>0.10$). Conversely, κ increased significantly with increasing BSD proportions at all C:SD levels ($p<0.01$) while E was not impacted by BSD at any clay level

(p>0.10). Importantly, these data, as well as the large variances observed within each filter type, highlight how input clay and sawdust proportions/sizes do relate with filter characteristics, though these relationships are not direct.

6.3.2 CWF Performance

6.3.2.1 Flowrate, Q

Shown in Figure 6.5 is the measured flowrate for each CWF disk aggregated by C:SD and BSD:SSD proportions. A secondary axis also illustrates pot-equivalent flowrates, which is the estimated flowrate that the same CWF disks would achieve if scaled to a CWF pot with Equation 6.4, adapted from Schweitzer et al. (2013).

$$Q_P\left(\frac{L}{hr}\right) = \frac{2\pi\kappa a}{h(n+1)(n+2)} \times z_0^{n+2} \qquad [6.4]$$

Where z_o is the height of water above the filter surface (i.e., hydraulic head) and a and n are scaling parameters estimated according to Equation 6.5. Using a CWF pot manufactured in the same factory as the disks, a and n were estimated as 2.89 and 0.49 with a standard error of 0.77%.

$$r = az^n \qquad [6.5]$$

Where r is the pot filter radius and z is the height from the filter base.

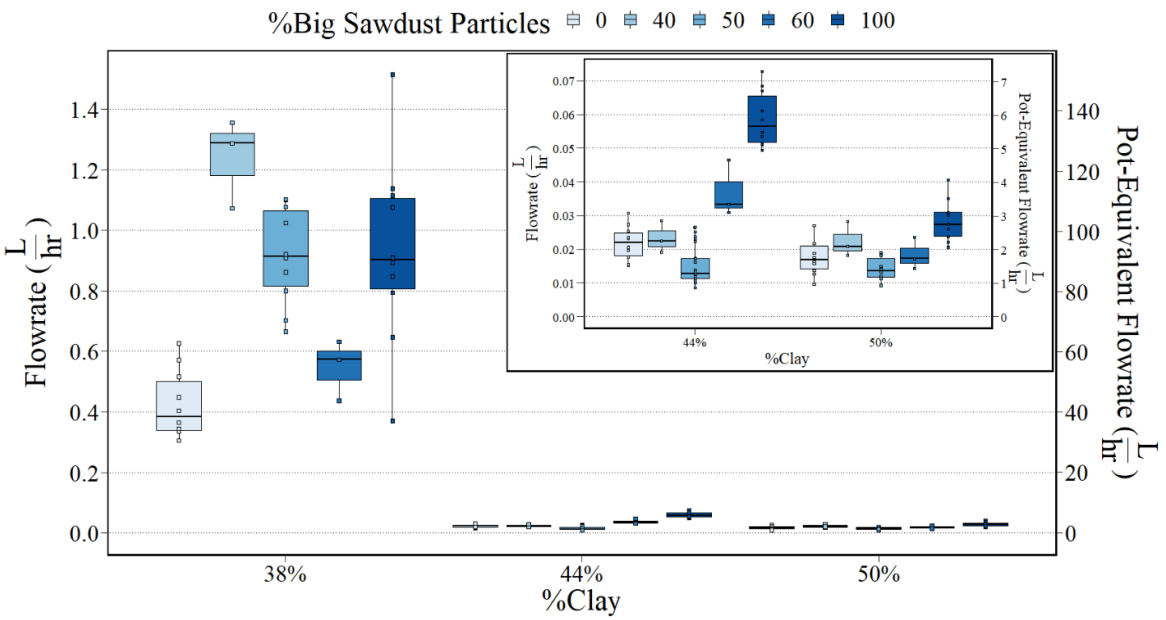


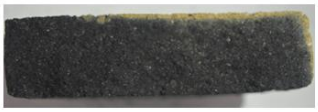
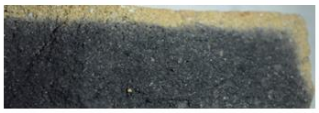
Figure 6.5 Measured CWF disk flowrates according to C:SD and BSD:SSD proportions. Inset: zoomed illustration of flowrates for filters with 44% and 50% clay proportions

Average filter flowrates were measured as 0.780 ± 0.324 L/hr, 0.028 ± 0.018 L/hr, and 0.020 ± 0.007 L/hr among filters with 38%, 44%, and 50% clay, respectively. All C:SD compositions were further significantly different from each other ($p<0.001$), yet on average, filters with 38% clay were 28 and 39 times faster flowing than those with 44% and 50% clay, respectively. Meanwhile, the impact of BSD proportion was also significant, though less drastic. For example, CWFs with 44% clay and 0%, 50%, and 100% BSD achieved average flowrates of 0.023 ± 0.004 L/hr, 0.014 ± 0.008 L/hr, and 0.058 ± 0.003 L/hr, respectively. Similarly, CWFs with 38% clay and the same percentages of BSD achieved average flowrates of 0.425 ± 0.109 L/hr, 0.916 ± 0.156 L/hr, and 0.931 ± 0.301 L/hr. CWFs with a larger nominal FM size thus achieved significantly larger flowrates than those with smaller FM sizes across clay levels. The overall trends were, however, indirect and nonlinear, the

reason for which is hypothesized to have emerged from changes in filter properties according to differences in internal heat transfer while firing in the kiln.

Specifically, FM combustion occurs at approximately 200-350 ºC, while complete burnout occurs at 700-800 ºC and clay vitrification occurs above 900 ºC (CMWG 2010). The way in which colour changes throughout the filter thickness is thus indicative of how it was fired and what temperature ranges were reached: a black colour is carbonous and suggests incomplete combustion and burnout (i.e., $T \leq 700$ ºC), while a beige-orange colour suggests clay vitrification (i.e., $T \geq 800$ ºC). The darker the orange colour, the higher the temperature that was reached. As such, evaluation of filter cross-sections in Table 6.4 clearly indicates differences in firing. Namely, filters with 38% clay are observably more orange than the other CWF types throughout the entire filter thickness, suggesting heat penetrated the filter body nearly uniformly. With that said, $C_{38}B_0$ filters had a somewhat blackened centroid while those with larger %BSD proportions did not. The temperature at the centroid therefore presumably did not reach the same level as those with bigger sawdust sizes. Similarly, $C_{44}B_0$ and $C_{50}B_0$ were mostly blacked throughout the entire disk thicknesses, whereas $C_{44}B_{100}$ and $C_{50}B_{100}$ were blacked only at the centroids (though more so in the former). Heat could therefore penetrate further towards the centroid when FM sizes were larger as well. These results align with previous research that has shown mean pore diameters may increase with increasing temperature exposure, resulting in significant increases in flowrate without increasing porosity or density by comparable margin (Soppe, et al. 2015, Rayner, et al. 2017).

Table 6.4. Filter firing by C:SD and BSD:SSD ratios

	38% Clay, 62% Sawdust	44% Clay, 56% Sawdust	50% Clay, 50% Sawdust
0% BSD, 100% SSD			
50% BSD, 50% SSD			
100% BSD, 0% SSD			

These changes are further supported by the theoretical principles of heat transfer through porous media. Namely, the temperature at any physical point, x, within the filter matrix during firing may be defined as $T(x) \sim f(d, t, \lambda)$ where d is the distance between the disk centroid and the disk surface, t is time, and λ is the porous material's thermal conductivity, defined by Collishaw and Evan's relation, $\lambda = \lambda_s\big(1 - \varphi(x,t)\big) + \varphi(x,t)\lambda_p$, where λ_s is the solid phase thermal conductivity, λ_p is the pore thermal conductivity, and $\varphi(x,t)$ is the porosity at the specific point, x, within the CWF matrix at time t (Smith, et al. 2013). In other words, heat transfer to the centroid is proportional to both the amount of pore space and solid material present between the surface and the centroid. As such, because CWF disks encounter heat radially, disk surface temperature is, by definition, higher than the centroid at t = dt and pore space is produced at the former before the latter. As time and kiln temperature then increase, FM near the surface burns out first and the associated increased porosity

facilitates increased heat penetration, leading to internal FM burnout. The observed relationship between filter colour and FM size and proportion can thus be understood accordingly. Similarly, differences between filters with the same input properties likely resulted from differences in encountered heat, and by extension, differences in burnout across the filter thickness. However, the CWF firing process remains highly understudied and additional research is required to elucidate how firing impacts filter characteristics and associated performance. Specific investigation of how internal filter temperature changes with time and space, as well as how those changes affect filter characteristics, is recommended.

6.3.2.2 Bacteria Removal, LRV

As shown in Figure 6.6, the differences in disinfection achieved by filters of differing input material parameters was notably less drastic than variations in flowrate. For instance, CWFs with all small sawdust particles (i.e., 0% BSD) and 50%, 44%, and 38% clay achieved average LRVs of 2.3 ± 0.4, 2.0 ± 0.4, and 1.3 ± 0.4, respectively. Similarly, CWFs with all BSD particles (i.e., 100% BSD) and the same clay proportions achieved average LRVs of 1.7 ± 0.3, 1.2 ± 0.2, and 0.6 ± 0.2, respectively. Increases in FM size and proportion thus led to associated decreases in LRV. The impact of FM size on LRV was, however, more significant among filters with 38% clay than others; average LRV for $C_{38}B_{100}$ filters was 117% that of $C_{38}B_0$, whereas the average LRVs of $C_{44}B_{100}$ and $C_{50}B_{100}$ filters were 67% and 35% larger than those of $C_{44}B_0$ and $C_{50}B_0$, respectively. Additionally, only filters with 44% and 50% were able to yield LRVs above the WHO guideline of 2 (%BSD-dependent). CWFs

with 38% clay, though far superior to the alternatives in terms of flowrate, were thus not acceptable for CWF production in the field. These observations support the prior hypothesis (section 6.3.2.1) that filters with 38% clay fired differently than others and resultingly yielded greater burnout and larger pore sizes than those with the same BSD:SSD ratios but less FM (i.e., more clay).

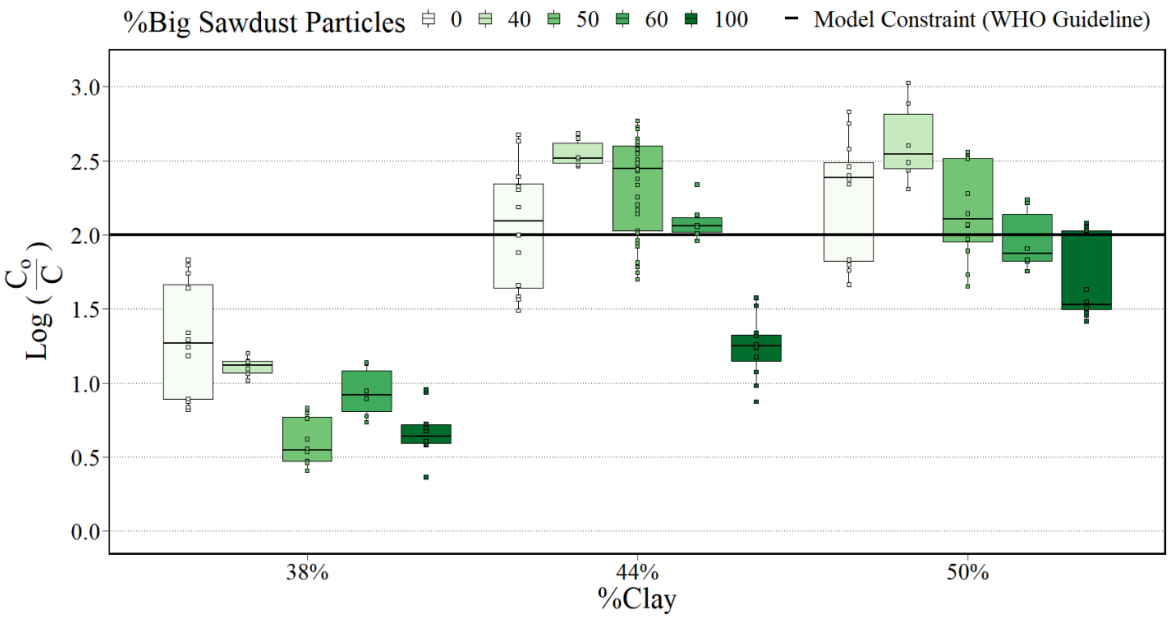


Figure 6.6. Measured CWF disk LRVs according to C:SD and BSD:SSD proportions

6.3.2.3 Filter Strength, MOR

Filter strength, as shown in Figure 6.7, was influenced by C:SD ratio, though not by the BSD:SSD ratio. On average, CWFs with clay percentages of 38%, 44%, and 50% achieved MORs of 0.7 ± 0.2, 1.3 ± 0.3, and 1.5 ± 0.4, respectively. Increasing clay content therefore led to increased strength, with greater impacts between filters with 38% clay and 44% clay ($p<0.001$) than those with 44% clay and 50% clay ($p=0.10$). A lower clay percentage than that used in the existing design may therefore yield an improved flowrate without

compromising strength. Meanwhile, omnibus ANOVAs between filters of differing C:SD ratios, as well as within each C:SD ratio highlight that %BSD is not a significant factor related to filter strength. Additionally, similar to observations for LRV, nearly all filters with 38% clay achieved MORs below the model inclusion criteria. These results thus further emphasize the inappropriateness of this C:SD ratio for field implementation. The wide variances observed within each filter type shown in Figure 6.7 further highlight how input material ratios are insufficient predictors of filter performance, and the utilization of more generalized parameters is needed.

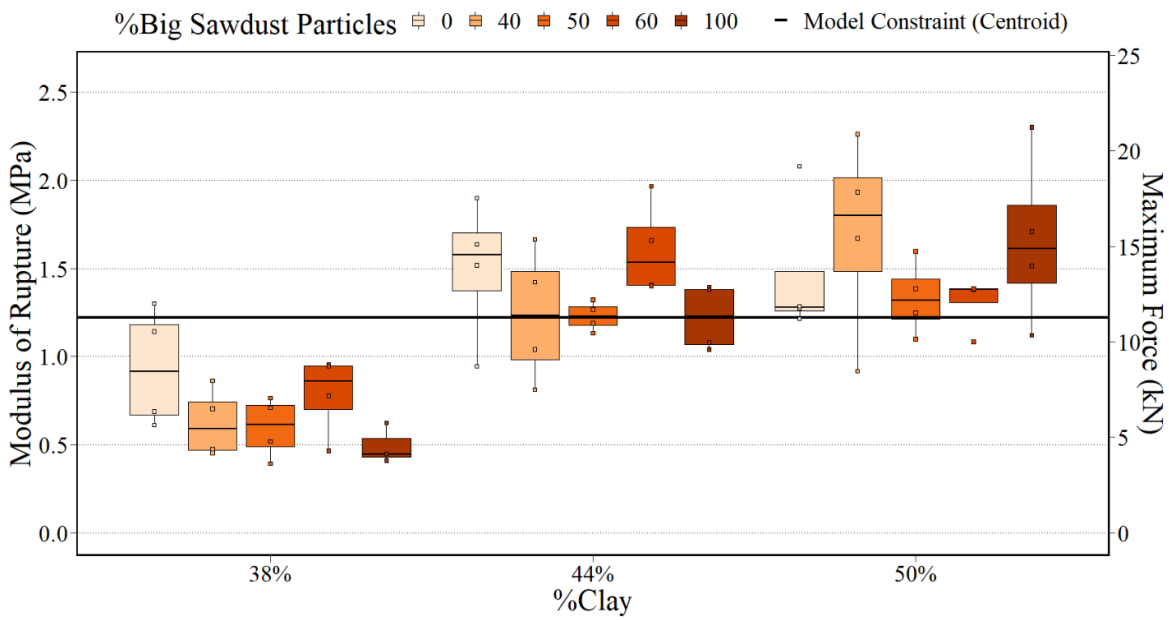


Figure 6.7. Measured Modulii of Rupture and Maximum Force (at fracture) for CWF disks according to C:SD and BSD:SSD proportions

6.3.3 Modeling

6.3.3.1 Model Fit and Accuracy

Shown in Table 6.5 are the final lower-level (i.e., input) and higher-level (i.e., output) models identified by the back-facing stepwise multiple regression analysis. All models developed using the nested structure were significant ($p<0.001$) and homoscedastic with a strong goodness fit and normally distributed residuals, as evidenced by the p-values for the Shapiro-Wilk coefficients and quantile-quantile plots in Figures 6.8A through 6.8F. Output models using C and BSD as predictors were, however, all heteroscedastic and thus invalid. Moreover, lower-level models were highly precise with standard errors ranging between 1.8 and 5.9%. The non-linear and interactive nature of clay proportion and sawdust size was therefore well captured by these porous media characteristics. CWF performance models were less precise though, even when using generalized predictors. For instance, the generalized LRV model had a standard error of 20.3%, nearly twice as large as those for Q and MOR, which were themselves nearly twice as large as the largest errors for the lower-level models. These observations illustrate how performance outcomes, and specifically LRV, are particularly challenging to estimate accurately. Further, they highlight that even though model outcomes were superior with general parameters, the present approach still faces significant limitations. That said, the value in using fuzzy logic for design optimization is exemplified.

Table 6.5 Stepwise-determined multiple regression models

Parameter	Model Equation	Adjusted R^2	Standard Error (%)	RMSE[1]	Shapiro-Wilk (p-value)
	Inputs				
Log Intrinsic Permeability, Log (κ)	$34.5 - 1.82\,C + 0.0192\,C^2 + 0.0172\,BSD - 0.000402\,BSD^2 + 0.00000308\,BSD^3 - 0.000102\,C * BSD$	0.949	1.79	0.165	0.110
Dry Density, ρ_D	$-6.26 + 0.320\,C - 0.00339\,C^2 - 0.005\,BSD$	0.800	5.24	0.058	0.456
Porosity, φ	$2.42 - 0.0765\,C + 0.000714\,C^2 + 0.00415\,BSD - 0.0000506\,BSD^2 + 0.000000338\,BSD^3 - 0.0000523\,C * BSD$	0.888	5.94	0.026	0.264
	Outputs - Nested				
Log (Q (mL/min))	$62.6 + 0.209 \times 10^8\,\kappa - 1.82\,\rho_D - 538.7\,\varphi + 1.78 \times 10^3\,\varphi^2 - 2.58 \times 10^3\,\varphi^3 + 1.42 \times 10^3\,\varphi^4 + 0.548 \times 10^8\,\kappa * \rho_D - 0.128 \times 10^8\,\kappa * \varphi$	0.971	11.8	0.117	0.101
LRV (Log (C_o/C))	$27.1 + 1.39 \times 10^8\,\kappa + 22.0\,\rho_D - 314.7\,\varphi + 851.4\,\varphi^2 - 651.5\,\varphi^3 - 2.45 \times 10^8\,\kappa * \varphi - 2.13 \times 10^8\,\kappa * \rho_D - 42.1\,\rho_D * \varphi + 3.69 \times 10^8\,\kappa * \rho_D * \varphi$	0.760	20.3	0.346	0.811
Log (MOR (MPa))	$3.79 - 2.70\,\rho_D - 8.55\,\varphi + 6.33\,\varphi * \rho_D$	0.685	10.32	0.107	0.466
	Outputs – Direct				
Log (Q (mL/min))	$42.2 - 1.81\,C + 0.0191\,C^2 + 0.0137\,BSD - 0.000413\,BSD^2 + 0.00000309\,BSD^3$	0.935	34.4	0.196	0.012
LRV (Log (C_o/C))	$-32.0 + 1.46\,C - 0.0154\,C^2 - 0.00510\,BSD - 0.000122\,BSD^2$	0.730	29.7	0.347	0.043
Log (MOR (MPa))	$-7.96 + 0.351\,C - 0.00378\,C^2 - 0.0104\,BSD + 0.000218\,C * BSD$	0.639	10.75	0.112	0.028

[1]RMSE = Root Mean Square Error

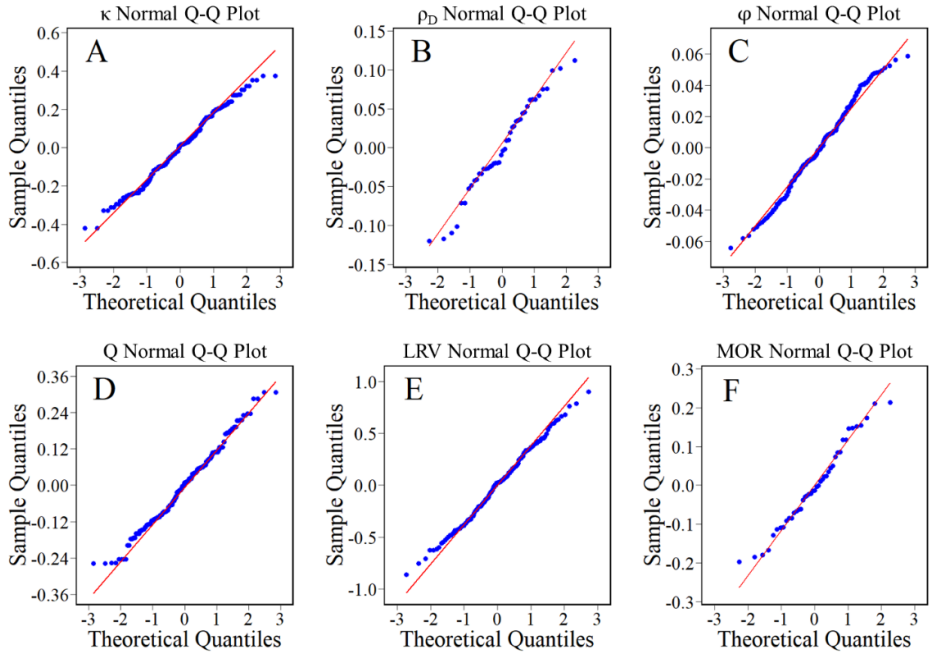


Figure 6.8 Normal quantile-quantile plots for regression models predicting (A) intrinsic permeability, κ (cm^2), (B) dry density, ρ_D (g/cm^3), (C) porosity, φ, (D) flowrate, Q (mL/min), (E) bacterial removal, LRV, and (F) strength, MOR (MPa)

How well the output models related to C:SD and BSD:SSD ratios may also be observed in Figures 6.9A-6.9I. Namely, average values for Log(Q) (Figure 6.9A), LRV (Figure 6.9D), and MOR (Figure 6.9G) were plotted in terms input manufacturing parameters. C:SD and BSD:SSD ratios that were not measured were then estimated using marching squares interpolation with the "plotly" package in R to produce the illustrated contours. The deterministic models for the same parameters using the nested structure, as shown in Table 6.5, were plotted adjacently in Figures 6.9B, 6.9E, and 6.9H, respectively. Additionally, the deterministic models for CWF performance that used %C and %BSD as predictors are shown in Figures 6.9C, 6.9F, and 6.9I. Each contour represents a difference of 0.1 units. As demonstrated, all deterministic models deviate from the measured contours, particularly when %C is 44% or higher and %BSD is less than 50%. Specifically, when %C and %BSD are used as model inputs, Log (Q) is underpredicted while LRV and MOR are both overpredicted therein. Conversely, nested models underpredicted LRV and MOR, while Log (Q) was underpredicted, but to a less significant degree. Further, changes in LRV are far less drastic with increasing %BSD when models were nested than when %C and %BSD were used directly. These results thus suggest the nested models provide more conservative predictions, adding strength to the confidence with which optimum estimates may be implemented. Additionally, the comparable shapes between models, particularly for Q and LRV, illustrates that the nested approach is valid and future research should investigate means of improving accuracy and precision with evaluation of filters from differing manufacturing contexts. With that said, the areas of superior and inferior accuracy highlight

how the deterministic estimates are insufficient for predicting CWF behaviours, emphasizing the value that fuzzy logic offers for narrowing the window of optimum filter design.

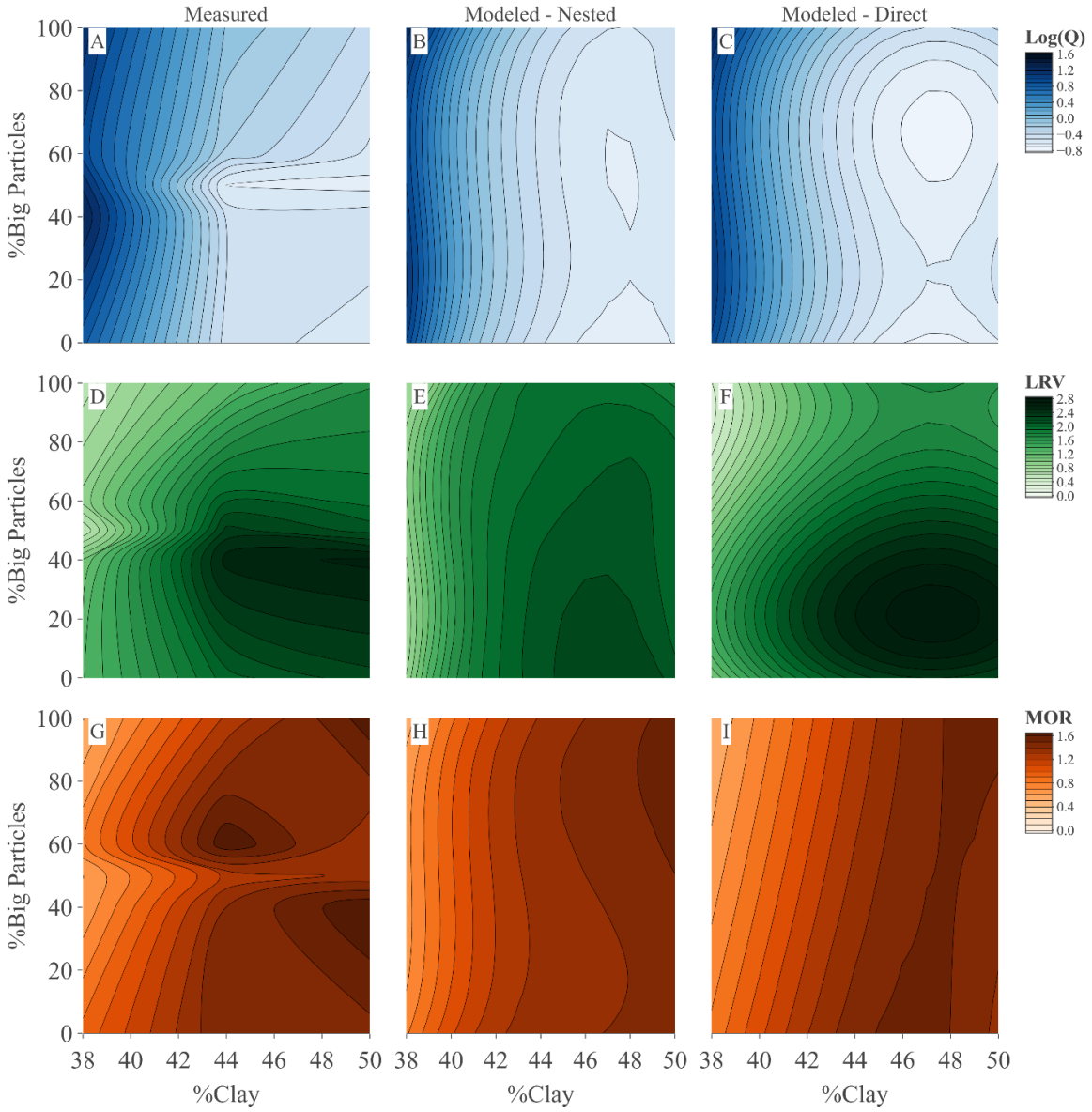


Figure 6.9 Contour plots illustrating the relationships between input material parameters and Log (Q) when (A) measured, (B) modeled with generalized predictors, and (C) modeled directly by C and BSD, LRV when (D) measured, (E) modeled with generalized predictors, and (F) modeled directly by C and BSD, and MOR when (G) measured, (H) modeled with generalized predictors, and (I) modeled directly by C and BSD.

A final critical note is that the conventional approach of estimating LRV from flowrate using a simple linear regression yielded an Adjusted R^2 of 0.59, standard error of 26.5%, RMSE of 0.444, and p-value for the Shapiro-Wilk test of 0.02. All measures of accuracy and precision were thus worse than those found for LRV when predicted with generalized parameters or C and BSD. This result confirms those from previous studies that the wide usage of flowrate as a proxy measure for bacteria removal effectiveness is inadequate (CMWG 2010, Schweitzer, Cunningham and Mihelcic 2013, Rayner, et al. 2017). A change in QA and QC measures and protocols is therefore recommended. Specifically, model performances emphasizes that, while still imperfect, utilization of generalized material characteristics for CWF performance prediction is superior to convention. CWF manufacturers should thus be encouraged to adopt such parameterization practices to predict performance outcomes.

6.3.3.2 Optimization Output

Shown in Table 6.6 are optimum estimates depending on the deterministic case, the safest-case, and the best-case scenarios. The deterministic case ignores the uncertainties and estimates the optimum material inputs and filter characteristics (i.e., κ, ρ_D, φ) from the nested models illustrated in Table 6.5 (i.e., correction factor = 1). The safest-case optimums represent the C:SD ratio, BSD:SSD ratio, and filter characteristics at which constraint requirements are always satisfied, even if the filter performance was the worst-case scenario of the model errors. Similarly, the best-case optimum represents the C:SD ratio, BSD:SSD ratio, and filter characteristics at which it is still possible that the constraints are satisfied

given model uncertainties. The presented Q's are the resulting optimum flowrates, calculated from the input material values at which constraints are satisfied for each individual case. Put another way, the range of values may be considered as possibility bounds within which constraint parameters may be satisfied, while Q is the resulting estimate at those bounds. Note that the possibility of failure increases towards the best-case optimums.

Table 6.6 Deterministic, safest-case, and best-case optimums for material inputs, filter characteristics, and filter performance.

Model Parameter	Deterministic Optimum	Safest-Case Optimum	Best-Case Optimum	Data Center Point
Material Inputs				
C:SD	42.2 : 57.8	47.3 : 52.7	40.3 : 59.7	44.0 : 56.0
BSD:SSD	33.0 : 67.0	0 : 100	0 : 100	50 : 50
Media Characteristics				
κ (x 10^{-9} cm^2)	16.72	3.606	28.72	1.809
ρ_D (g/cm^3)	1.167	1.337	1.165	1.301
φ (%)	47.64	39.20	48.08	43.20
Performance Outcomes				
Q_{DISK} (L/hr)	0.042	0.013	0.083	0.015
Pot-Equivalent Q (L/hr)	4.15	1.28	8.20	1.56

The PBDO model estimates that filters produced with clay percentages ranging from 40% to 47% and BSD percentages ranging from 0% to 33% may yield full CWF pots with flowrates ranging between 1.3 and 8.2 L/hr while still satisfying bacteria removal and strength requirements. When generalized, these results suggest that filters should have a density ranging between 1.16 and 1.34 g/cm^3, a porosity between 39% and 48%, and an intrinsic permeability between 3.6 x 10^{-9} and 28.7 x 10^{-9} cm^2. Both the safest-case and best-case model scenarios also predict optimums with only small SD particles, though clay proportion optimums are markedly different. This result suggests that, as also illustrated by van Halem et al. (2017) and others, flowrate may be increased without compromising disinfection by

increasing the percentage of FM within the CWF fabrication mixture while keeping FM size small.

6.4 Limitations and Future Work

While this research has laid a valuable foundation for CWF optimization, several study limitations exist. For instance, the sawdust sizes evaluated herein were limited in scope and do not represent the full range of options available. They were also relatively uncontrolled in that the size distributions were large due to the partner factory's method of sieving in parallel rather than in series. Future work should therefore evaluate CWF performance using smaller sizes with narrower ranges to advance progress towards a truer optimum filter design. Similarly, the nested models produced within this research were developed, in part, to facilitate scalability between facilities using differing clays, firing materials, and firing processes by predicting performance according to characteristics instead of input parameters. However, all filters in this research were produced by a single factory, meaning the models are tuned to the processes therein. Additional work is therefore required to confirm the models' applicability in diverse settings. Finally, the significant observed model variability may have resulted from the artisanal and consequently chaotic nature with which CWFs are manufactured. As such, given this embedded uncertainty within the fabrication process, it is possible that model accuracy and precision would improve with additional data. Future work should therefore build upon this framework to strengthen the models with the inclusion of additional data such that convergence towards a more precise optimum may be achieved.

6.5 Conclusion

The objectives of this research were to (1) evaluate the impacts of varying FM size and proportions on filter flowrate, bacteria removal, and strength, (2) evaluate the use of generalized characteristics of porous media as predictors of key filter performance metrics, and (3) determine an optimum range of filter material parameters that maximize flowrate while maintaining acceptable levels of bacteria removal efficacy and filter strength. Results demonstrate that both FM size and proportion significantly impacted flowrate and disinfection, whereas strength was only impacted by proportion. Further, the use of generalized parameters for filter performance modelling proved an effective and valuable approach that may afford CWF manufacturers superior capacity with respect to QA and QC procedures.

Other key outcomes include:

- Optimum CWF κ, ρ_D, and φ should range between 3.61×10^9-16.7×10^9 cm^2, 1.17-1.34 g/cm^3, and 39%-48%, respectively to maximize flowrate and maintain LRV above 2 and MOR above 1.2.
- Flowrate alone offers inferior predictive capacity of disinfection efficacy than when κ, ρ_D, and φ are used in combination
- PBDO found optimums had smallest sawdust sizes, suggesting larger FM proportions with smaller sizes may lead to improved flowrates with acceptable bacteria removal when compared to existing CWF design (i.e., center point of CCD).

- Deterministic modelling is unreliable for predicting CWF performance without accounting for embedded uncertainty.

Additional research is required to investigate model applicability across fabrication contexts, specifically within the optimum ranges identified in Table 6. Future work should also evaluate smaller FM sizes with more narrow ranges than those used in this study.

6.6 Acknowledgements

We would like to thank the Basu Research Group, and specifically Rachel Boyer for her specific contributions to the production of the present datasets. We also wish to thank Wine to Water International and the staff at Safe Water Ceramics of East Africa who produced the filters used in this work and have supported us throughout the entirety of this study. This research was funded by the Natural Sciences and Engineering Council of Canada (NSERC), and Carleton FutureFunder.

Chapter 7: Towards a Participatory Framework for Improving Water & Health Outcomes: A Case Study with Maasai Women in Rural Tanzania

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Abstract

Rural communities in sub-Saharan Africa (SSA) are disproportionately burdened by a pervasive lack of access to safe drinking water. Widespread programmatic failure in the water, sanitation, and hygiene (WaSH) sector has resulted in particularly slow progress in alleviating these challenges in the region. Drawing from decolonial and participatory methodological scholarship, this research demonstrates how geographically- and demographically specific, locally controlled, and long-term educational programming can improve health and wellness outcomes when associated with a technological intervention. Specifically, consultations between January 2015 and August 2018 were followed by an iterative and community-driven program development process between January and July 2019. Fifty Maasai women were subsequently recruited to participate and were provided with a point-of-use water treatment technology in August 2019. These women engaged in a series of three 14-week WaSH education programs over an 18-month evaluation period. Results showed that 38% of participants reported regular diarrhea at baseline, decreasing to 8%, 0%, and 3% immediately after each of the three WaSH education programs were

provided at 3, 12, and 18 months. Interim measurements taken between WaSH programs showed 35% of participants (at 6 months) and 5% of participants (at 15 months) reporting regular diarrhea. A trend of improvement was thus observed over the study period, though the increase in reported diarrhea at 6 months demonstrates the need for long-term commitment on the part of WASH practitioners when engaging with end users to achieve sustained change. Further, this research highlights the importance of participatory program development and pedagogical approaches in WaSH interventions, where local control of study objective determination and implementation, combined with consistent and long-term engagement, can facilitate sustained technology use and associated reductions in diarrhea.

Keywords: Ceramic water filters, Community engagement, Diarrheal Health, Gender, Participation, WaSH, Water treatment

Highlights:

- Codeveloped participatory framework for WaSH intervention through community interviews
- Integrated feminist and decolonial theories into program structure
- Intervention resulted in less filter breakage than earlier studies.
- Diarrheal health and recall of lessons improved with time and education provision.
- Results show WaSH programs must be long-term and locally conceived and implemented

7.1 Introduction

The pervasive lack of access to safe drinking water remains among the most pressing global challenges, affecting over 2 billion individuals of whom more than half reside in rural sub-Saharan Africa (SSA) (World Bank 2021). Diarrheal illness resulting from unsafe water consumption is a central factor in an annual death toll of almost 500,000 children under the age of five years, of whom 73% are in SSA (UNICEF 2019). In addition, water insecurity and chronic diarrheal episodes have been linked to malnutrition and undernourishment (Brown, Cairncross and Ensink 2013, K. Brown 2003), conditions that adversely impact both economic development (Heltberg 2009, Dadson, et al. 2017) and overall mental and physical well-being (Nguyen, et al. 2014, Bolton and Robertson 2016). Climate change is further exacerbating these problems, as increasing frequency and intensity of storm events facilitates more rapid transmission of communicable diseases (Dias, et al. 2018), and warmer temperatures with associated worsening droughts create higher contaminant loads due to reduced dilution capacities (Bates, et al. 2008). Similarly, the COVID-19 pandemic has illustrated the critical role that water, sanitation, and hygiene (WaSH) all play in managing health, especially as this particular virus is easily transmitted by vectors such as hand touching and faecal-contaminated water. Rural communities in which such services are most limited are thus those also most at risk of becoming further marginalized. Significant improvements in WaSH conditions are urgently required.

7.1.1 Background: Water Access in Rural sub-Saharan Africa

Progress in rural geographies has, however, been particularly slow. For instance, only 12% of rural populations in SSA have access to safely managed drinking water services compared with 50% of their urban counterparts. Rural populations in SSA have further only observed a 6% increase in safe water service access since 2000 compared with a 15% improvement in rural areas worldwide (World Bank 2021). And though there is certainly a diversity of reasons for this disparity, widespread programmatic failure is undoubtedly a contributing factor. For example, a 2018 systematic review of WaSH intervention studies conducted in low-income countries found that between 69% and 93% of participants failed to sustain usage of a water treatment technology after only 6 months (Martin, et al. 2018). Knee et al. (2021) further found "no evidence that the intervention affected the prevalence of any measured outcomes after 12 or 24 months of exposure" when evaluating a non-governmental organization's introduction of toilets to 495 households in Malawi, where only the technology itself was provided. Similarly, Marshall and Kaminsky (2016) estimated that 40% of all WaSH interventions overall fail to achieve their intended results, a rate that has remained stagnant since the 1980's. Thus, while economic investment and broader practical constraints may hinder the ability to implement programs (Alaerts 2019, Njoh, Ananga, et al. 2018) and require attention, implementation and engagement strategies must also be carefully conceived to ensure long-term success.

There has been wide discursive recognition that the introduction of technology alone to a given geography does not necessarily result in health improvements (Bishoge 2021, UNICEF 2016). A growing field of research has thus highlighted the need to facilitate behavioural changes and nudge individuals towards proper WaSH practices (e.g., handwashing,

elimination of open defecation, treatment of drinking water) to gain improvements in WaSH outcomes (Fiebelkorn, et al. 2012, Dreibelbis, et al. 2013). Yet, effective strategies to achieve such WaSH-related behavioural change (WBC) and associated health outcomes remain uncertain given the vast intra- and inter-community diversity across SSA and beyond. In fact, efforts to develop a standardized WBC methodology may be impractical; each geographic location and the individuals residing in it have unique characteristics and are motivated by different factors. In other words, WBC and associated technology adoption cannot be uniformly scaled. Rather, contrary to the generalized concept of "scaling-up" WaSH programming, disaggregated and locally defined approaches oriented towards specific target populations may be the most appropriate methodology for mobilizing change (C. P. Sianipar, et al. 2013, Marshall and Kaminsky 2016). United Nations Sustainable Development Goal 6.1.b states that the provision of WaSH services must therefore involve "community participation and engagement" to ensure longevity (UN 2021). However, the translation of *participation* as a concept into an appropriate and effective practice that produces intended health and wellness outcomes over an extended period remains unelucidated, hindering progress in the WaSH and WBC fields.

7.1.2 Gender and Colonization Among the Maasai of Tanzania

Gender and its relationship with colonization constitutes an additional dimension of the discourse surrounding WaSH inequities and their associated challenges. The relevance of gender to water and health in SSA, and particularly how water-related responsibilities are disproportionately burdensome for women, is widely accepted (CSDH 2008, Masanyiwa, Niehof and Termeer 2014, R. Brown 2010). Yet seldom do sectoral discourses appreciate

the way colonialism and its legacy contributed to, and continue to reinforce, this inequity. For example, the imposition of water collection and management responsibilities on women and girls is not the problematic, but rather that such responsibilities have produced a gendered power imbalance (Hellum, Kameri-Mbote and Koppen 2015). The role of colonialism in producing this imbalance is thus of critical importance to understanding why such conditions persist, as well as how they may be overcome. In the case of Maasai women in rural Tanzania, the group and geography central to this intervention (see Sections 7.1.3 and 7.2.1), such impacts are clearly observable.

Before British colonization, Maasai were not a single homogenous group but rather a diverse set of semi-nomadic peoples connected by a shared lingual history ('Maasai' literally translating to 'speakers of the Maa language'). When Tanganyika came under British control in 1916, these disparate communities were confined and constricted within what was termed the 'Maasai Reserve' and managed as a singular 'tribe' (D. Hodgson 2001a). Historically accessible natural resources were thus subsequently limited, particularly as land appropriation for agricultural, mining, and tourist activities accelerated (Bruner and Kirshenblatt-Gimblett 1994, Mbilinyi 2016, D. Hodgson 2001a). Historical forms of social organization were also transformed according to imported European economic values and biologist (i.e., organization according to sexual difference) labour prescriptions (Agbaje 2021, Oyewumi 1997).

Pre-1916 labour among Maasai was non-gendered and divided according to primarily practical considerations. Animal husbandry, water retrieval, and other daily tasks were often completed by men and women interchangeably, though the former was more commonly a

male endeavour when women would to tend to children or address health concerns (Saitoti 1988). Yet, British occupation produced two concurrent phenomena. First, 'household' tasks such as cleaning, cooking, childcaring, health treatment, retrieving firewood and water, or milking livestock became considered inferior to the work of cattle keeping and grazing when administrators suggested animal husbandry's explicit economic importance meant it was superior (Rigby 1992c). Further, the colonists imparted the belief that women should not participate in financial matters, or even own anything themselves, and thus categorized cattle management as an exclusively male enterprise in which women should not participate (D. Hodgson 2001a, 2001c). This imposed labour valuation undoubtedly contributed to the disenfranchisement, and often violence, that continues today (Pommells, et al. 2018, D. Hodgson 2001a).

Colonial land appropriation and associated Maasai mobility restrictions also created additional labour burdens for women. Whereas pre-reservation Maasai nomadism was defined largely by the availability of water and grazing lands (Rigby 1992c, D. Hodgson 2001a), restrictions resulted in resource reductions that increased travel times for retrieval, increased health burdens, and further removed women from the newly identified areas of economic importance. As colonial occupation transitioned into post-colonial nationalism and capitalist structures became ubiquitous, Maasai sedentism was normalized and associated power and labour inequities were concretized (R. Brown 2010, D. Hodgson 2001c, Mbilinyi 2016). These labour inequities are also continually worsening as climate change increases the distances required to retrieve water (particularly during dry seasons) and rapid urbanization draws resources from rural areas (Bates, et al. 2008, Dadson, et al. 2017). The

contemporary sexual discrimination of labour organization, the related sexualized structures of power, and the hardened physical requirements of household maintenance are all moreover borne of exclusionary colonial policy, maintained by the ubiquity of capitalism, and continue to intensify with its associated socioeconomic and environmental effects (D. Hodgson 2001c, Oyewumi 1997, Kurtis, Adams and Estrada-Villalta 2016).

Guidance from local Maasai gender rights advocates from the present community has indicated that to erode these historical power structures, it is necessary to ensure the demonstration of female participation in material ownership and positions of social authority. Further, reducing labour burdens was identified as a critical component of accelerating gender equity so that women could have available time to engage in income-generating activities. Based upon this consultative framework, the research objectives were set to instill a sense of material ownership for the women, reduce labour burdens associated with unsafe water consumption, and build capacity and confidence among the female program stewards and the participants themselves to facilitate a sense of gendered empowerment. See Section 7.1.3 for further detail.

7.1.3 Study Concept and Objectives

This study integrated a locally engaged and participatory approach to the promotion of improved WaSH-related behaviours and associated health outcomes among Maasai women and their children in rural Tanzania. Drawing from decolonization scholarship, the intervention was created with the intention of actively addressing the effects of development and public health practices in the colonial period that continue to manifest in persistent

inequities today (Chouinard 2016, Richardson 2019, Kurtis, Adams and Estrada-Villalta 2016). Per the detailed effects (Section 7.1.2), it was explicitly designed to mitigate embodied risks produced by environmental marginalities (i.e., consumption of unsafe water); to ensure that local knowledge systems were not diminished by the promotion of Westernized health-related behaviour; and to situate decision-making, implementation, and determination of health objectives under local control. The aim was to study a model for meaningful and inclusive participation in a WaSH program, from development to implementation, that encourages local ownership, promotes desired behaviours through locally relevant and culturally specific methods, and ultimately facilitates self-determination with regard to drinking water.

In this particular intervention study, point-of-use (POU) ceramic water filters (CWF) were provided as a water treatment technology for improving drinking water quality. As shown in Figure 7.1, CWFs are pot-shaped filter elements used in the home that treat water through combined physical and chemical bacterial removal processes (Venis and Basu 2020). Previous research has further shown that CWFs are among the most effective home-based treatment technologies, as well as those most preferred by users (Wolf, et al. 2014, Burt, et al. 2017, Santos, Pagsuyoin and Latayan 2016). CWFs were thus distributed and implemented alongside an in-depth WaSH education program designed to align with Maasai ontology and modes of production (i.e., social and economic practices). Associated health outcomes were measured and assessed before intervention, as well as at multiple time points during the evaluation period (see Section 7.2.3).

Figure 7.1 Ceramic Water Filters (from left to right): after manufacture, being used in a field setting, and being tested for flow without their encasing bucket.

7.2 Methodology

7.2.1 Community Details: Kimokouwa, Tanzania

This research was conducted within the Kimokouwa ward of the Longido District in the Arusha Region of northern Tanzania, located approximately 15 km north of the central Longido Village, the economic hub of the district (see Figure 7.2a). Kimokouwa has an estimated population of approximately 10,000 inhabitants with a density of about 24/km^2, illustrating its remote nature (Brinkhoff 2012, Worldometer 2020). The vast majority of inhabitants identify as Maasai and practice modes of economic activity such as livestock keeping and beadwork. They typically live in Maasai homesteads called *bomas,* which are widely dispersed across the ward in sub-villages. Note that bomas are grouped living quarters where multiple homes (Figure 7.2b) are circularly oriented and enclosed by acacia-branch fences (Figure 7.2c).

The local climate is semi-arid (Figure 7.2d), with only about 350 mm of precipitation per year, nearly 75% of which falls during the first wet season (from February to the beginning

of April). Consequently, water in the village is collected predominantly from communal taps (Figure 7.2e) that are fed by water collected in basins (Figure 7.2f) located on the slopes of neighbouring Mount Longido). During the wet season, some water is also collected from streams fed by mountain runoff, from rainwater, or from a shallow well borehole. During the dry seasons, water is often collected from engineered dams (Figure 7.2g) or dugout wells (Figure 7.2h).

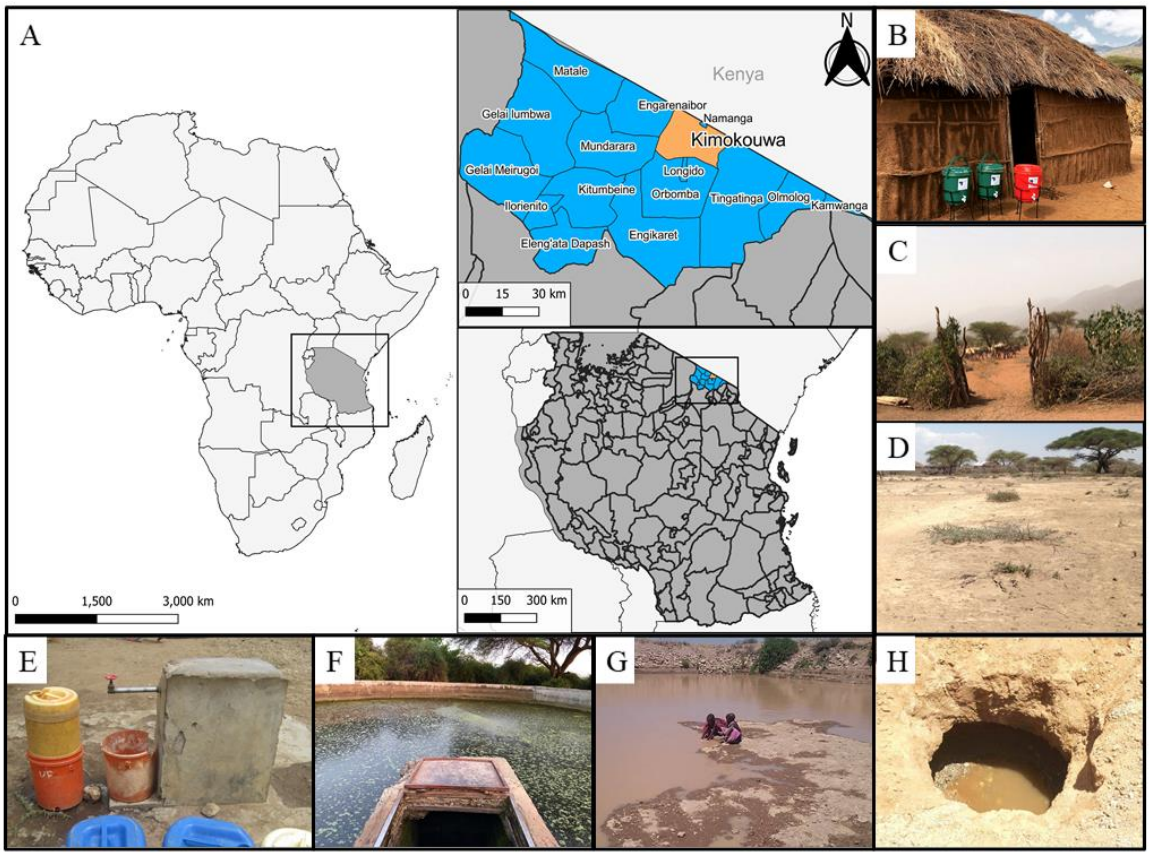

Figure 7.2 (a) Map of Tanzania (Inset: Longido District with Ward Labels). Data from Humdata.org (Humanitarian Data Exchange 2018). (b) A Maasai house within a boma in Kimokouwa, with three CWFs in front. (c) Entrance to a traditional boma. (d) Dusted plains of Kimokouwa during the dry season. (e) A communal tap for collecting water. (f) A basin atop Mt. Longido from which tap water comes. (g) An engineered earthen dam with children collecting water. (h) A dugout hole from which women collect groundwater

7.2.2 Program Development

Engagement in the Longido District was initiated by an invitation from community leaders who wished us to attempt to address challenges they had identified related to water resources and water-related health outcomes. Preliminary consultations took place between January 2015 and August 2018 and were focused on (1) gaining geographically specific information on what water sources were being consumed; (2) how water was managed; (3) what cultural and social institutions exist within the community; and (4) how those institutions affect the lives and livelihoods of local residents. In answering the first two questions, 663 female heads of households in Kimokouwa were briefly interviewed by local research assistants using door-to-door recruitment. In addition, while utilizing interactive consultation methods, undergraduate students and staff researchers from Carleton University met with local leaders to identify specific areas of interest and present potential options for addressing them. The latter two questions were answered through what McCoy (2012) describes as a *methodology of encounters*, in which no specific recruitment procedure is followed, and no desired outcomes are specified at the outset. Significant time was rather spent with a variety of individuals across the district, ranging from secondary school students to healthcare workers to Maasai cattle keepers to bead workers, where all interactions were informal and undirected.

This methodological choice marked the first step towards a community-driven approach, as the determination of the key challenges to address was based on local identification rather than a Western-oriented prescription. Familiarizing ourselves with the community members and becoming familiar to them during initial consultations also acted as a trust building

exercise that facilitated the ease by which future community and financial resources could be mobilized.

After initial consultations were completed, the program development process occurred between January and July 2019. This process also drew on theories of decolonizing research methods. Key objectives, participant interaction methods, interview questions, and all other factors of the program implementation were established through iterative communications with key local informants. Specifically, desired learning objectives identified by members of the Longido community during the consultation phase were first cross-referenced with WaSH education resources freely provided by the World Health Organization (WHO) and the Centre for Affordable Water and Sanitation Technology (CAWST), allowing for the determination of critical information for inclusion in the program, as well as which metrics were to be used for evaluations (WHO 2020, CAWST 2020). An initial program design was then developed and subsequently discussed in detail with staff and volunteers at the Tanzanian Education Micro Business Opportunity (TEMBO), a non-governmental organization based in Longido, Tanzania, and the primary implementation partners and collaborators in this research. The program design was then updated according to their feedback, and this process was repeated until key informants' approval was obtained. A similar process was subsequently completed with key informants from Wine to Water (W2W) and Wine to Water East Africa (W2WEA), this projects' industrial partners, as well as nurses at the Longido District Health Clinic (LDHC). A final program was then presented to village leaders and the head doctor at the LDHC for approval. All interview questions, education resources, and program scheduling may be found in Appendix C. This research

was approved by the Carleton University Research Ethics Board B, the Tanzanian Commission for Science and Technology, and local representatives of TEMBO (an NGO registered under the Tanzanian NGOs Act, 2002).

Please note that all CWFs used in this study were produced by W2WEA, a locally owned and operated social enterprise based in Arusha, Tanzania (70 km from Longido), which is supported by W2W, a non-governmental organization (NGO) located in Boone, North Carolina, USA.

7.2.3 Program Implementation and Structure

Fifty women attending TEMBO's Adult Literacy Program in Kimokouwa were recruited into the study and individually asked to provide informed consent before participating. Upon receipt from every group member, we provided a general overview of the project structure and objectives and answered any questions from the group, including questions about us as individuals to facilitate interpersonal familiarization. Education programming began in the week following this activity, during which participating women were provided with a pictorial education pamphlet on WaSH-related topics. This pamphlet was used to guide 14 weekly sessions provided by our local research assistants, "P" and "T" (see Appendix C). Note that P and T are Maasai women under 30 years of age who were born, raised, and educated within the Longido District. Both were recruited through an openly advertised hiring process initiated by TEMBO in which applicants were required to meet specified qualifications including an ability to speak English, Kiswahili, and Maasai, past experience working within a community setting, and some knowledge of how to use a smart phone. Both

were subsequently trained by [author; removed for peer-review process] in data collection, interview methods, participatory pedagogical methods, and general WaSH information before beginning interactions with program participants.

After the third education session, a CWF and filter stand (Figure 7.3) were delivered to the individual homes of every woman in the group, and boma members of all genders were instructed to respect the filter as the property of the participant. Community leaders further reinforced this notion after agreeing to assist in managing power dynamics associated with technology management during the program design phase.

Figure 7.3 Ceramic water filters in use in participant homes

Education was provided in a participatory format, where information was relayed through discussion and question-answer style dialogue (Figure 7.4). The detailed learning objectives were divided into 7 different lessons, which were each completed twice consecutively (i.e., Lesson 1 in both weeks 1 and 2, etc.) within the 14-week program. This repetition schedule was decided upon to ensure that every woman was present for at least one provision of a lesson, as women commonly missed meetings for a diversity of reasons; this decision was further taken on the recommendation of TEMBO's community facilitator and Adult Literacy

Program Coordinator, [author; removed for peer-review process]. Each one of these lessons lasted approximately 15-30 minutes in duration. The entire 14-week program was also repeated three times (see Figure 7.5), aligning with an agile approach to meeting participant needs recommended by previous health research among Maasai women in Tanzania (Birks, et al. 2011).

Figure 7.4 "P" conducting education with Maasai women in Kimokouwa, Longido, Tanzania

7.2.4 Data Tracking, Management and Analysis

Structured interviews were conducted before intervention, as well as at approximately 3, 6, 12, 15 and 18 months from the time of filter reception. All interview materials were translated into both Kiswahili and the Maasai language, to facilitate complete understanding of questions being asked. Though a 9-month follow-up was originally scheduled, restrictions on meetings due to the COVID-19 pandemic prevented this from occurring. Interview data was collected using the mWater smartphone app under W2W's secure licence. Data was then

downloaded locally and analyzed using RStudio. Note that due to the self-reported nature of the health data and logistical limitations on water quality monitoring, filter and education implementation effects were analysed together rather than separately.

One participant was provided a filter at baseline but did not complete an initial interview, three participants dropped out of the study by the time of the 3-month interviews, one more dropped out by the time of the 6-month interviews, and six interviews could not be conducted at the 6-month mark due to COVID-19 related restrictions. Further, due to other diverse and significant challenges produced by COVID-19, literacy group attendance decreased drastically after its resumption, resulting in only 16 of the initial 50 women in the study being interviewed at the 12-month mark. Attendance increased thereafter and 39 participants were interviewed at the 15-month mark and 36 were interviewed at 18 months (three participants were unreachable at this time). No selection bias was observed in the 12-month results, as outcomes among those present were consistent with the rest of the group at other measurement points. Therefore, though participation at this time was low, extrapolation of the inferences remains appropriate.

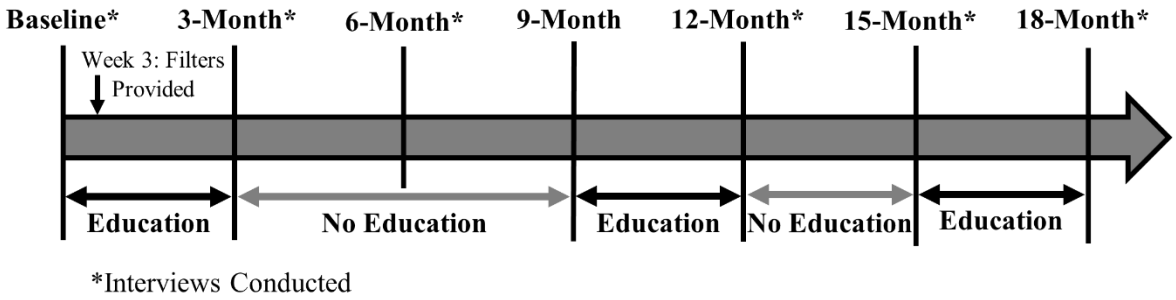

Figure 7.5 Project Timeline

7.3 Results

7.3.1 Initial Consultations

7.3.1.1 Kimokouwa Site Evaluation

Preliminary interviews in Kimokouwa found that the average household was comprised of 5.2 ± 1.9 (one standard deviation) people with an average of 2.6 ± 1.5 children under twelve years of age. Of those households, 69% reported using less than 80 litres of water per day and 31% reported using less than 40 litres per day, which approximately equate to the WHO limits of 20 litres per person per day to "realise minimum essential levels for health and hygiene", and 7.5 litres per person per day to meet a person's needs in terms of "survival, basic hygiene, and basic cooking", respectively (Reed and Reed 2013). Further, 90% of households also reported having 2 or less buckets or barrels to store their water (i.e., < 40 L of storage capacity). Given the reported amount of water used described above, one can infer that most women venture to collect water more than once per day, which is a significant burden in terms of both time and labour (an observable product of colonially enforced sedentism; see Section 7.1.2). The self-contained design of the CWF, which stores up to 10 L of filtered water at a time, is therefore a particularly beneficial element of the technology for these households.

7.3.1.2 Lessons from Encounters

Consultations with local stakeholders produced a diversity of information that was useful for the development of our WaSH program. Information about local attitudes towards water and health proved particularly informative. For example, conversations with various residents

illustrated that diarrhea is not widely considered an issue among community members but is rather an unavoidable characteristic of the Longido lifeworld. Nearly every interviewee reported either having cholera or typhoid at some point in their lives or reported having an immediate family member who had suffered such diseases. Few, however, associated these diseases to unsafe water consumption or poor sanitation. As put by one interviewee when speaking about her home village:

> *This is our life, fetching [water] from a tap or bucket and putting it to the mouth to drink. We didn't see any problem. It was, like, the way we live so we didn't see any problems. […] Diarrhea? Yes! Of course! Mostly kids. But we did not know the problem was maybe related with water until we would go to the hospital or the clinic where you get some knowledge or education and they would say 'do this, do that.'*

Discussions with local health authorities further revealed that many of the challenges they face in addressing water-related illnesses in the community come from a persistent belief in a relationship between spirits and stomach health. With most Maasai in the community following a hybrid of some monotheistic faith denomination and traditional spirituality, people commonly hold strong beliefs that they regularly interact with the divine and that some form of higher power directly contributes to their circumstances. In keeping with the tenets of the decolonizing research methods literature, we let these findings significantly inform the education program; we attempted to carefully navigate these beliefs to ensure that no conflict was perceived between them and the scientific information being shared by local program staff.

Consultations also revealed that several interviewees considered existing local water treatment solutions as undesirable and expressed specific interest in CWFs as an alternative. For instance, many community members noted their dislike of available chlorine solutions due to an unfavorable added taste, no removal of suspended solids, and required regular purchasing that created significant long-term cost burdens. Chlorine availability was also noted as a challenge due to inconsistent supply, whereas the CWFs were considered superior in this regard due to their being manufactured in a nearby city. Furthermore, respondents reported knowledge of options such as boiling water, letting water sit in charcoal, or purchasing bottled water, but expressed disinterest in all due to time and/or cost implications. Presumably due to CWFs having none of these limitations, both community leaders and broader members expressed interest in their trial after presentation by the researchers.

7.3.2 Home-Use Filter Program

7.3.2.1 Study Participant Demographics

As shown in Table 7.1, participant ages ranged between 16 and 55 years, with the majority reporting an age between 25 and 35. Similar to the results from the site evaluation, an average of 2.3 ± 1.0 children per household was also reported, 47% of whom were reported to be under 5. As such, approximately 210 individuals are estimated to have access to a filter at the time of study inception. It is also noteworthy that 44% of participants were unaware of their household monthly income, highlighting the prevalence of female exclusion from areas of economic importance among this group. Further, of those who could report their income,

88% reported incomes below the Tanzanian national poverty line (TZS 49,320 per month per adult equivalent) (World Bank Group 2021).

Table 7.1 Sociodemographic Details of Participants

Variable	Categories	n	% of Group
Age	16-25	12	24%
	25-35	20	40%
	35-55	13	26%
	55+	5	10%
Marital Status	Married	37	74%
	Single	6	12%
	Not Answered	7	14%
Educational Achievement	Never Attended School	30	60%
	Primary School	8	16%
	Secondary School	1	2%
	Not Answered	11	22%
Children Ages	<5	53	48%
	5-10	36	33%
	10+	21	19%
Religion	Christian	38	76%
	Other	12	24%
	Not Answered	10	20%
Income Source[1]	Cattle Keeping	46	92%
	Market Selling	13	26%
	Other	4	8%
Income	Lower Quartile (20000 TZS[2])	7	14%
	Lower-Middle Quartile (32000 TZS)	6	12%
	Upper-Middle Quartile (60000 TZS)	6	12%
	Upper Quartile (200000 TZS)	6	12%
	Don't Know[3]	22	44%
	Not Answered	3	6%

[1] Some participants had multiple sources of income, so percentages do not add to 100%.
[2] Tanzanian shillings in 2019.
[3] "Don't Know" refers to participants who do not have access to their household finances.

7.3.2.2 Filter Breakage

Filter breakage is widely recognized as one of the most common causes of filter disuse (Roberts 2004, CMWG 2010). In the present study, seven filters (14%) broke by the 3-month interviews, increasing to nine (18%) by the 6-month interviews, eleven (22%) by the 12-month interviews, and twelve (24%) by the 15-month interviews. No additional filters broke

between 15 and 18 months. The participants with broken filters also attended an average of 5.0 ± 2.6 of the original 14 education classes compared with 9.3 ± 3.6 among those with unbroken filters, demonstrating a significant relationship ($p<0.01$) between class attendance and filter care. In addition, these results are superior to those observed in literature; for example, Brown (2007), Roberts et al. (2004), and Lemons et al. (2016) found 45%, 25%, and 18% of distributed filters broke within 44, 11, and 1.5 months, respectively. The present education program thus likely translated to comparatively improved filter care. With that said, the filter breakage observed in this study and others suggests that some breakage is inevitable and is a condition for which WaSH practitioners and interveners must plan.

7.3.2.3 Diarrheal Illness

The self-described diarrheal outcomes reported at each survey phase of the study are shown in Figure 7.6. Diarrhea fluctuated between measurement times: 38% of participants reported having diarrhea with some frequency at baseline, changing to 8% at 3 months, 32% at 6 months, 0% at 12 months, 5% at 15 months, and 3% at 18 months. A significant difference in diarrheal outcomes was thus observed after the second (9-month) and third (15-month) education sessions were completed, suggesting a single 14-week program was insufficient to facilitate improvements. Additional interfacing and long-term programming, however, increased the intervention effectiveness.

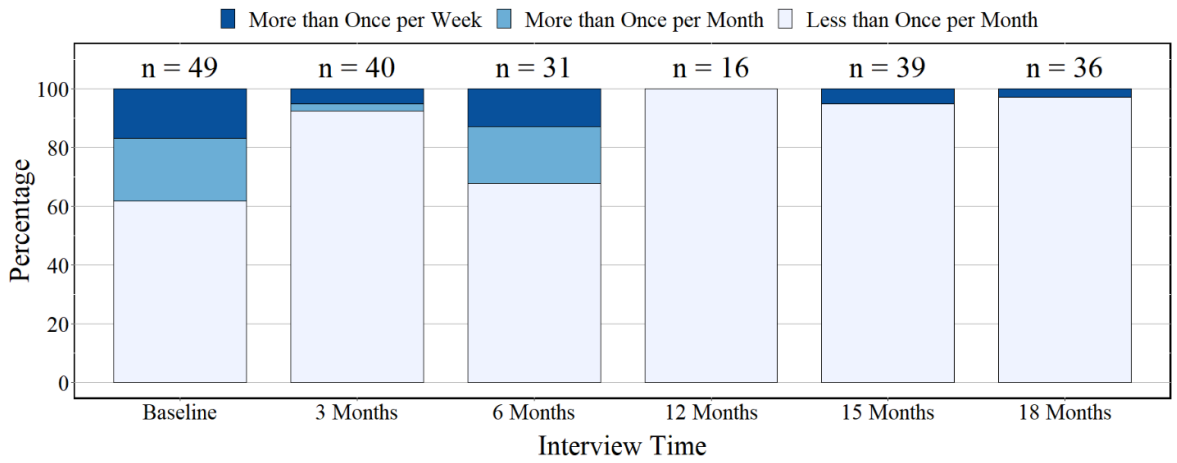

Figure 7.6 Reported Diarrheal Outcomes Among Participants at Each Survey Phase.

Diarrheal outcomes among children were lower at baseline than previous research had estimated, though still approximately three times higher than the Tanzanian national average (Mshida, et al. 2017, Edwin and Azage 2019). The trend after intervention, as reported by their mothers in Table 7.2, was also slightly different than the participants themselves. Specifically, 19% of children were reported to experience some level of diarrhea at baseline, changing to 2%, 20% and 5% at 3 months, 6 months, and 12 months, respectively. At the 15- and 18-month measurements, however, diarrheal occurrence increased to 17% and 30%, respectively. This observed increase is particularly concerning when age is considered: of the children reporting diarrhea at 15 and 18 months, 65% and 83% were under the age of 5, respectively. However, diarrheal severity among participating children did improve after intervention. Because filter performance monitoring was outside the scope of this research, it is impossible to say with certainty if this change was a result of filter failure, transmission by other vectors, or both. In other words, diarrheal outcome is an inherently incomplete metric of CWF effectiveness. This notion is further particularly acute among children who typically spend more time away from the home than their mothers and thus may encounter

pathogens via different communication pathways. Scrutiny of participants' chosen water source, however, suggests filter failure may be at least partially relevant.

Table 7.2. Diarrheal frequency of Participants' Children, Aggregated by Age

Survey Phase	Age	Diarrheal Frequency					
		Less than Once per Month		More than Once per Month		More than Once per Week	
		n	(%)	n	(%)	n	(%)
Baseline	<5	37	(34)	10	(9)	6	(5)
	5-10	31	(28)	3	(3)	2	(2)
	10+	21	(19)	0	(0)	0	(0)
3-Month	<5	50	(47)	0	(0)	0	(0)
	5-10	37	(35)	0	(0)	0	(0)
	10+	17	(16)	1	(1)	1	(1)
6-Month	<5	24	(28)	2	(2)	0	(0)
	5-10	17	(20)	5	(6)	0	(0)
	10+	28	(32)	9	(10)	1	(1)
12-Month	<5	12	(33)	1	(3)	0	(0)
	5-10	12	(33)	0	(0)	0	(0)
	10+	10	(28)	1	(3)	0	(0)
15-Month	<5	40	(31)	15	(11)	0	(0)
	5-10	40	(31)	4	(3)	0	(0)
	10+	27	(21)	3	(2)	1	(1)
18-Month	<5	26	(25)	26	(25)	0	(0)
	5-10	34	(32)	5	(5)	0	(0)
	10+	14	(13)	0	(0)	0	(0)

Shown in Table 7.3 is the conditional probability of consuming water from a given water source and reporting illness at each reporting time ("More than once per month" AND "More than once per week"), calculated per Equation 7.1.

$$P(Source|Illness) = P(Source) * P(Illness) \quad [7.1]$$

where P(Source|Illness) is the conditional probability of consuming water from a given source and reporting illness, P(Source) is the probability of consuming water from a given source, and P(Illness) is the probability of reporting illness when having drunk from a given source.

At baseline, the data shows only marginal differences in the correlation of illness with consumption of water from a well, village tap, surface water, or dam water source. The consumption of rainwater proved least likely to have caused illness. Data collected before intervention therefore did not provide any clear indication that one specific source water influenced diarrhea over others, other than a lower prevalence of diarrhea occurring with consumption from rainwater collectors. After intervention, however, clearer differences may be observed. Namely, at 6-, 15-, and 18-month reporting times, when diarrheal cases were highest, there was a disproportionately high probability of reporting illness if one consumed village (untreated) tap water relative to the other sources: 11.6%, 8.2% and 25.5%, respectively. Village tap and well water in Kimokouwa also typically have lower turbidity (dissolved and suspended particles) than other sources, which is a water quality factor that has been linked with filter performance: higher turbidity leads to higher levels of microbiological pathogen removal (Abebe, Chen and Sobsey 2013, Farrow, mcBean and Salsali 2014). As such, it is possible that contaminant loads were higher during these periods than others, which were removed less effectively by the filters challenged with clearer water. Future research quantifying filter performance and source water contaminant loads over time is therefore needed. With that said, people encounter a multitude of vectors for disease, especially children who spend time outside the home and do not always drink filtered water. Further, diarrhea is known to fluctuate with changing weather, as well as form in spatial clusters; illness commonly begets more illness. Temporal variations in reported health were thus likely influenced by factors other than only drinking water quality, meaning these observations must be considered additive rather than exclusively explanatory.

Table 7.3. Conditional Probability of Drinking Water from a Source and Reporting Illness

Water Source	Reporting Time					
	Baseline	3-Month	6-Month	12-Month	15-Month	18-Month
Well	6.8%	0.8%	0.0%	0.0%	0.3%	0.0%
Tap	8.4%	1.2%	11.6%	1.8%	8.2%	25.5%
Surface	7.0%	0.8%	4.8%	0.0%	1.4%	1.3%
Rain	0.8%	1.6%	6.3%	0.0%	0.0%	0.0%
Dam	3.8%	1.2%	3.7%	1.8%	7.2%	0.7%

7.3.2.4 Co-Factors of Health

In addition to assessing health directly, the evaluation of participants' retention of key learning objectives from the education sessions offers insight into whether the associated behaviours changed accordingly and thus whether risk of illness from non-water vectors was reduced. For instance, as shown in Figure 7.7, 58% of participants could recall more than 3 of 6 specified times for handwashing at baseline, which increased to 70% after the first education session was provided. After the first pause in education sessions, that value dropped to 42% at the 6-month period, then increased to 74% after the second education session was provided from the 9th to 12th months. Finally, retention increased to 100% at the 15- and 18-month marks. Like the diarrheal health outcomes, a general trend of improvement over time was observed. Thus, the longer people were engaged in the program and the more education they received (particularly after the second session), the better retention became and the more impactful, theoretically, the education program was on health outcomes.

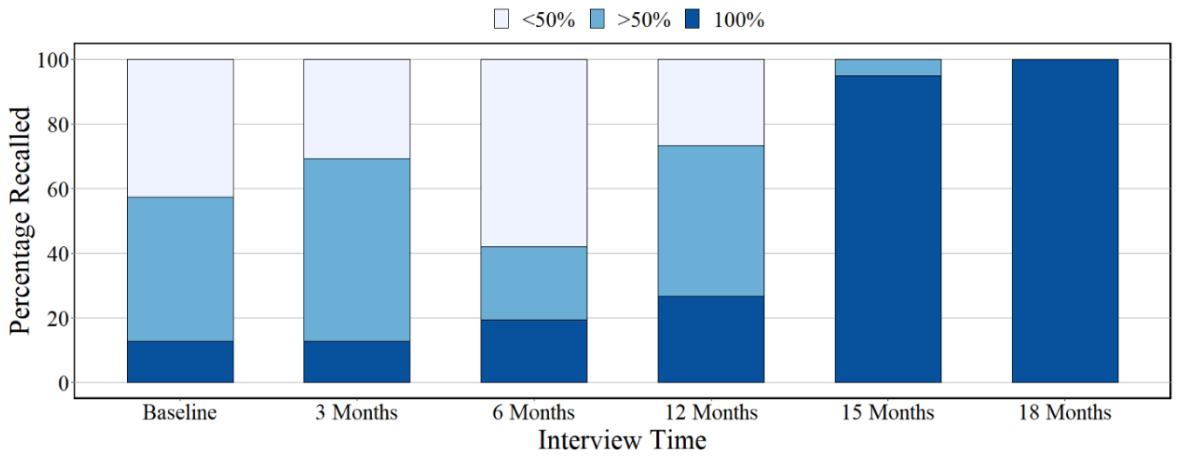

Figure 7.7. Retention of learning objectives on when it is appropriate to wash one's hands. Given times include: (1) after defecating, (2) before eating, (3) before feeding a child, (4) before preparing meals, (5) after cleaning a baby, (6) after milking animals.

Similar observations were made after analysis of participants' recollection of the filter maintenance and safety protocols reported at each time point (Table 7.4). Like health and handwashing, outcomes fluctuated with amount of education received, with a general positive trend towards complete retention with time and education repetition. When summing all 14 of the maintenance protocols provided, 90% of participants could recall at least 50% at the 3-month interviews (after the first education session), decreasing to 36% at the 6-month reporting time (three months after the first pause in programming). Retention then increased to 67% after the second education session at 12 months, improving thereafter to 100% at both the 15- and 18-month reporting times. It thus appears that continual engagement and the repeated provision of information directly targeting specific behaviours, in combination with the provision of solutions and support to facilitate behavioural change, produced positive results across numerous dimensions.

Table 7.4 Participants' recall of filter maintenance protocol at each survey phase

Indicator	Response	Survey Phase									
		3 Months		6 months		12 months		15 months		18 Months	
		n	(%)	n	(%)	n	(%)	n	(%)	n	(%)
Filter Maintenance Rules Recalled[1]	<50%	4	(10)	18	(58)	4	(25)	0	(0)	0	(0)
	>50%	5	(13)	13	(42)	3	(20)	1	(2)	0	(0)
	All	28	(72)	0	(0)	8	(50)	38	(97)	36	(100)
	NA[3]	2	(5)	0	(0)	1	(7)	0	(0)	0	(0)
Filter Cleaning Steps Recalled[2]	<50%	1	(2)	10	(31)	3	(19)	0	(0)	0	(0)
	>50%	4	(10)	20	(63)	4	(25)	2	(5)	0	(0)
	All	36	(85)	1	(3)	7	(44)	37	(95)	36	(100)
	NA	1	(2)	0	(0)	2	(12)	0	(0)	0	(0)

[1] (1) use a clean cup, (2) no playing near the filter, (3) be gentle with the tap, (4) do not touch the tap opening, (5) do not touch the outside of the filter, (6) leave the filter in the bucket (unless cleaning), (7)leave the filter in the stand (unless cleaning)

[2] (1) clean the bucket and lid with soap and water, (2) scrub the inside of the filter with a brush and boiled water, (3) remove grimy residue with boiled water and dump, (4) scrub the outside of the filter with brush and boiled water, (5) pour boiled water over entire filter, (6) enclose filter within cleaned bucket and lid, (7) remove all water from lid before turning right-side-up

[3] NA = No Answer

Though the education program also provided participants with information on vectors for WaSH-related illness such as open defecation and the sharing of water supplies with livestock, it did not provide participants with any associated intervention methods or capacity for change related to these two elements. Unsurprisingly then, as noted in Table 7.5, no change in behavior was observed, with open defection remaining at initial reporting level of approximately 83-84% and sharing water with livestock remaining at 81-82%. Yet, when asked if the water they poured into the filter (including that which may be shared with animals) was clean, a clear change can be observed after the second education session was provided at the 12-month mark: while 75% considered it clean between baseline and 6 months, 9% considered it clean between 12- and 18-month marks. Together, these data highlight that knowledge transfer of the importance of safe WaSH practice alone is insufficient to mobilize efforts to overcome material constraints. The affordability and/or

availability of material, in combination with improvements in understanding of their value, is therefore critical to mobilizing substantial and meaningful change.

Table 7.5 Participant Responses to Health Co-Factors Not Targeted by Intervention

Indicator	Response	Survey Phase											
		Baseline		3 Months		6 months		12 months		15 months		18 Months	
		n	(%)	n	(%)	n	(%)	n	(%)	n	(%)	n	(%)
Sharing Water with Animals	Yes	40	(82)	33	(82)	20	(65)	9	(56)	37	(95)	33	(92)
	No	9	(18)	6	(15)	11	(35)	6	(38)	2	(5)	1	(3)
	NA[1]	0	(0)	1	(3)	0	(0)	1	(6)	0	(0)	2	(6)
Defecation Location	Toilet	6	(12)	7	(18)	5	(16)	3	(19)	6	(15)	0	(0)
	Open Defecation	41	(84)	32	(80)	25	(81)	10	(63)	33	(85)	35	(93)
	NA	2	(4)	1	(2)	1	(3)	3	(19)	0	(0)	1	(3)
Perception of Water Source	Clean	30	(61)	31	(78)	29	(94)	3	(19)	5	(13)	0	(0)
	Unclean	17	(44)	8	(20)	2	(6)	12	(75)	34	(85)	36	(100)
	NA	2	(4)	1	(2)	0	(0)	1	(6)	1	(2)	0	(0)

[1] NA = No Answer

7.4 Discussion

The results indicate that this intervention included several successful elements, as well as limitations that require further exploration. For instance, the marked reduction in diarrheal outcomes observed over time clearly demonstrates that this CWF intervention greatly reduced incidence of water-related illness. An observed trend towards complete retention of the concepts communicated in the educational interventions, as well as a decreasing rate of filter breakage over time and increasing class attendance, also indicate that observed improvements in health may be attributed not only to the provision of filters, but to the education program as a whole. In other words, as cognitive understanding of actionable WaSH practices improved (e.g., handwashing) and participants took greater care of their filters, associated health improved accordingly. Because these changes were strongest after more than one 3-month education program was provided, we may conclude that

technological interventions should be accompanied by long-term, relevant, and appropriate information provision activities. One-off or short-term technology and/or WaSH education provision may be insufficient to produce meaningful changes in health compared with repeated, continual, and culturally specific engagement.

The observations among children, however, demonstrate inherent limitations to this study. The fact that, overall, 38% and 10% of participants reported illness before and after intervention, respectively, while 19% and 15% of children reported illness, highlights that the program was more challenged in improving child health outcomes than adults. As there are diverse vectors by which individuals, and children in particular, may encounter WaSH-related diseases, these observations demonstrate that a singular approach to their mitigation is inherently limited. Programs of this nature should therefore integrate diverse and complementary initiatives related to water, sanitation, and hygiene to substantially reduce the risk of illness across a community landscape. Similarly, programs must include water quality monitoring to isolate sources of illness.

Another important consideration is that, though filter breakage was indeed significantly reduced when compared with observations from the literature, the fact that filter breakage occurred at all highlights an important limitation of POU technology. That is, regardless of the degree of behavioural change achieved, there is a significant probability that some filters will break. The occurrence of such predictable accidents (e.g., livestock knocking over a filter after entering the home) should not, however, define whether an individual has access to safe water n the long-term. Future research into POU technology, and CWFs specifically, must therefore look beyond WBC alone and investigate models for alleviating the burden of

care from the individual so that the risk of breakage would be reduced further than what was practically feasible in this work. Additionally, the cost and ease of replacing the technology must be such that access to filtered water is not prevented by practical and financial constraints when filters do break.

Finally, aside from the broad implications for WaSH interventions, these outcomes also provide important insights regarding progress towards gender equity and Maasai environmental self-determination. Most significantly, the group meeting structure framing this intervention offered critically important space for gender-focused community building. TEMBO has cultivated an environment in which marginalized women can congregate and organize to better their lives, while also being able to behave informally and discuss topics that may otherwise be taboo. The organization has created a powerful space that actively counteracts the continuing impacts of labour biologism, which alienates women, and by extension, limits their resistance (Oyewumi 1997). Because labour contributions by Maasai women have been trivialized and solidified according to colonial value prescriptions, opportunities for discussion around means of overcoming these circumstances are necessarily limited. Group meetings centred around knowledge sharing offer space for discussion and self-improvement are often unavailable elsewhere. Further, the program's development and implementation was driven by the participants' peers, thus embedding locality within the initiative. The knowledge base from which participants could advocate for their own well-being with water was couched in community relevance. And by including the WaSH discussions within TEMBO's broader program on human rights, lingual and financial literacy, and other matters, the study demonstrated an approach to building WaSH

into the larger landscape of initiatives necessary for mobilizing progress towards equity and justice. As this work is ongoing and participants will continue to be interviewed over time, future research will involve identifying how this program contributes to future local water-related policy and priority setting.

7.5 Conclusions

This research paper detailed a decolonizing and participatory methodological approach to improving safe water access and associated health outcomes among a rural community of Maasai women and their children in Longido, Tanzania. Several important observations made in this study highlight various ways in which WaSH practices and policies may be improved within the rural SSA context. Namely, a key finding is that the long-term, community-driven, and participatory engagement strategy employed in this study might be usefully incorporated into future programs and expanded further. For example, the utilization of a robust consultation process ensured the intervention was able to target the challenges identified as most acute within the community, reducing the influence of Western biases. The orientation of the education program around Maasai praxis and epistemology further demonstrably increased understanding of mitigating WaSH-related health risk; as put by one participant, "I have lived with stomach pain for my entire life, but now it has stopped. I didn't know it was from the water." By ensuring that the lessons were taught in a respectful and culturally specific manner, and that they were taught by individuals with whom participants could identify, this approach offered a useful framework for improving the likelihood of participant retention of information. Additionally, the development of local capacity through training and hiring of local project facilitators was an important step in ensuring that

knowledge would grow and remain within the intervention area over time. The emphasis on building local knowledge and an aptitude for program development and implementation thus assists in producing longevity and encourages self-determination. Finally, ensuring that the provision of information was responsive to participants' and facilitators' needs, that project facilitators were regularly available for support, and that users knew there was a program infrastructure available to help them if needed, demonstrably resulted in successful outcomes over time. The relationship between technology users and WaSH quality/health evaluators should thus be consistent and long-term. It is further recommended that, where possible, future research evaluate how engagement of this sort could be expanded to encompass more features of the WaSH landscape to enable individuals to access safely managed water and sanitation services simultaneously.

Chapter 8: Behaviour Change in International Water, Sanitation, and Hygiene: A 100-Years Perspective

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Submitted to Journal of International Studies Perspectives (Under Review)

Abstract:

The current methodological paradigm for addressing water, sanitation, and hygiene (WaSH) inaccessibility in rural sub-Saharan Africa (SSA) is achieving insufficient progress. This essay evaluates WaSH-related policy, programming, and discourse from 1918 to 2021 to identify how this paradigm evolved and how it may reform. I argue that political-economic environments have strongly influenced existing sectoral praxis, shaping both programmatic methods and means. Namely, colonial occupations generated rural-urban material inequalities, which were maintained and exacerbated during post-war reconstruction (1950-1970) and the proliferation of neoliberalism (1970-1990s). Meanwhile, modernization theory, a primary driver of colonialism, has persisted discursively and practically. That is, in response to rural resource limitations, WaSH practitioners promoted lower-cost *appropriate technologies* in the 1980s. Then, with challenges regarding technological disuse and misuse, behaviour change oriented methodologies responsively emerged into the 2000s and continue today. Yet, much like colonial predecessors, this latter turn presupposes that its programmatic benefactors must adapt to access WaSH services. Behaviour change programs thus fail to critically consider the role of technological inadequacies and associated risk

exposures in perpetuating existing inequities. Investigation of utility-style service models, where WaSH services adapt to the lives of its benefactors and behavioural persuasion is substituted for non-user technological management, is recommended.

Keywords:

Colonialism, Rural Water, Sub-Saharan Africa, WaSH, WaSH Methodology

8.1 Introduction

The pervasive lack of access to safe drinking water, sanitation, and hygiene (WaSH) remains among the most pressing global challenges of the 21st century. More than 2 billion people are estimated to lack access to either safe water, sanitation, or both, most of whom live in rural Sub-Saharan Africa (SSA) (World Bank 2021). WaSH is further regarded as the single greatest cause of disease burden (GBD Risk Factor Collaborators 2020), with its effects on physical health, mental health, livelihoods, and overall wellbeing having shown to produce social, political, economic, and environmental marginality that hinders progress towards justice and equity more broadly (Clemens and Douglas 2012, Nguyen, et al. 2014, Bolton and Robertson 2016). With climate change, COVID-19, and diverse other crises exacerbating inequities and their influences on lived conditions, understanding how to alleviate these burdens is urgent.

Various scholars have posited different approaches to achieving WaSH equity, though significant attention within the sector has most recently been directed towards behavioural models (Fiebelkorn, et al. 2012, Dreibelbis, et al. 2013, Bishoge 2021, WHO; UNICEF 2017). Facilitating the adoption of so-called *proper* WaSH behaviours (e.g., regular handwashing, using and maintaining water treatment and/or sanitation technology) is widely considered a necessary feature of any program in the Global South (Jamison, et al. 2006). As put by the United Nations Children's Fund (UNICEF), "the importance of encouraging improved hygiene behaviours [has become] entrenched in sector programming and plans" (UNICEF 2016). Similarly, Bishoge (2021) states in a systematic review of sectoral methodological trends that among other factors, "people's behaviours were identified as the

main constraints to sustainable sanitation and hygiene in [Sub-Saharan Africa].” Vast epidemiological literature further demonstrates the profound relevance of individual behaviours in the transmission of microbially-induced illness, suggesting this focus too aligns with modern scientific evidence and models for disease causation (Martin, et al. 2018).

The effectiveness of this approach is, however, highly debated. While some argue “the occurrence of specific behaviours is all that is needed to prevent disease” (Ginja, Gallagher and Keenan 2021), others have demonstrated that facilitating these changes in behaviour is notoriously challenging (Middleton, Anton and Perri 2013). For example, Huda et. al (2012) found less than 3% of participants among 850 households in Bangladesh washed their hands with increased frequency 18 months after intervention, and Knee et al. (2021) found “no evidence that the intervention affected the prevalence of any measured outcomes after 12 or 24 months of exposure” when evaluating a non-governmental organization’s (NGO) introduction of toilets to 495 households in Malawi. Tamas and Mosler (2011) further found nearly 70% of participants either stopped using or never started using solar disinfection water treatment 7 months after a 2-month promotion period in Bolivia, and summarily, a meta-analysis by Martin et al. (2018) found between 65-93% of participants in SSA failed to change WaSH behaviours within 6 months of program implementation. Why, then, does WaSH-related behaviour change (WBC) remain so central to sectoral policies and programming? How has it emerged as a key mechanism for alleviating water-related burdens and what are the implications of its persistence?

8.2 What is WaSH Behaviour Change?

WBC methodologies are based on epidemiological literature that states washing hands, drinking clean water, using toilets, and other common '*good'* WaSH practices improve health (Jamison, et al. 2006, Dreibelbis, et al. 2013). The theory is responsive to insights from germ theory, which states that microbial contaminants are nearly ubiquitous and are communicated between individuals according to several vectors: e.g., eating, shaking hands, drinking water. WBC is therefore an intersectional field between psychology, public health, and engineering that intends to reduce the risk of disease transmission by ending certain behaviors during which it is most likely to occur. The foundational premise of this field is that individual behaviours are fundamental determinants of microbiological disease, and it is these behaviours which, if changed through targeted through policy and praxis, can reduce WaSH access inequities. Identifying the best methods for facilitating this behavioural change is further among the largest subfields of the WaSH sector, with dozens of theories currently under evaluation (Fiebelkorn, et al. 2012, Kraemer and Mosler 2010).

WBC critics suggest the kernel of the problematic does not lie with the individuals but with the socioeconomic and political system within which inequal circumstances manifest. The causal pathway is not person-to-person communication, but centuries of structural violence that have led to conditions in which the risk of transmission is higher in lower-income nations than wealthier ones. From this perspective, WaSH-related illness results from rational decisions made by individuals on how to meet their overall needs given excessive resource restrictions and limited adequate solutions for alleviating burdens. It is not due to a lack of knowledge or understanding. As such, critics argue that WBC methods commit "epistemic

violence" by virtue of "hermeneutic injustice", operationalizing "malfeasance in the interpretation of observational and measured data" to perpetuate cross-national North-South hegemony (Richardson 2019, Pohlhaus 2012). The foundation of this critique is that inequal economic and policy environments facilitate the manifestation of inequal risk, making them the fundamental determinants of microbial disease. Public health engineers and WaSH practitioners would therefore best service intended benefactors by developing a novel framework for implementation that places the burden of alleviating epidemiological risk onto the state and international institutions/implementers rather than the individual (Ray and Smith 2021). Put another way, the environment surrounding the individual must change, not the individual themself.

Indeed, the essence of the WBC debate is one of structural versus individual responsibility. Do individual behaviours scale towards global challenges, or do global superstructures manifest as challenges at the individual level? And what is the relationship between discourse, policy, economics, and praxis? This research review aims to illuminate this debate by providing a detailed account and analysis of the technical, social, political, and economic dimensions of WaSH sectoral history within the context of international development in rural SSA. The primary objective is to disentangle the respective roles that structural resource inequities, individual behaviours, and international WaSH discourse play in the production and treatment of regularized disease transmission in these contexts. A history of WaSH from 1918 to 2021 will be presented first, with particular attention placed upon rural SSA and how WBC emerged in these geographies. A discussion of this analysis will follow, with

recommendations of how policies and practices may be reformed to meet Sustainable Development Goal (SDG) 6, universal safely managed WaSH services, by 2030.

8.3 Methods

References used herein were chosen via a snowball method. Keyword searches on SCOPUS were completed to identify a list of sources for each time period individually. A reduced list of key texts were then identified according to their relevance and appropriateness to the present discussion, as determined by the author. Referenced sources therein were subsequently used to provide additional context and examples as needed. Specific attention was further placed on sources from, or about, major international institutions including the World Bank and United Nations because of their positions as key centres of international discourse. Each evaluated publication was finally coded categorically according to (1) time period, (2) context (e.g., research, policy, program), (3) topic (e.g., implementation, history), and (4) geography (rural, urban, country) and used throughout this writing accordingly.

Case studies were also chosen according to the following criteria: (1) implemented an intervention related to water, sanitation, hygiene, or a combination; (2) implemented by an international institution or organization; (3) occurred during the time period of discussion; (4) was implemented within rural SSA.

8.4 A Brief History of WaSH

8.4.1 WaSH and 20th Century Colonialism (1918-1948)

The end of World War I (WWI) was chosen to begin this discussion because it marked an important shift in the organization of exogenous political power. Namely, the 1919 Treaty

of Versailles led to British and French confiscation of German overseas colonies, allowing for an expansion of their global empires and an increased concentration of power. Concurrently, the discourse surrounding what was an appropriate use of that power had begun to shift. The 1917 Bolshevik revolution, for instance, heralded a new wave of anti-imperialism that opposed any supranational subjectification, especially when primarily for economic extraction (Darwin 2009). United States President Woodrow Wilson's Fourteen Points Speech in January 1918 also detailed American opposition to European annexation of overseas geographies and emphasized the need to transition colonies towards national self-determination (Manela 2007). Fear of an expanded people's movement, fear of uprisings in the colonies if populations were not rewarded for their war efforts, and an interest in maintaining US political support led both French and British colonial administrations to change their strategies for overseas governance (Kitchen 2017). Consequently, subjects thereafter would yet not be able to realize self-determination or political power, but certain populations were newly permitted to work in lower levels of the colonial administrations and benefit from public services (Aminzade 2013, Darwin 2009).

International development, as its known today, has furthermore been highlighted by various historians as unofficially beginning at the Great War's end. That is, it was mostly thereafter that colonial administrators created, curated, and implemented programs intended to specifically benefit the wider colonized populations (Lorenzini 2019a, 9-21, Horner 2020, D. Hodgson 2001a, 48-92). WaSH development in SSA may also be considered as having begun at this time, as waterworks were too guided by external actors to benefit some locals among the colonial administrators and select elites (Huillery 2009, Njoh and Akiwumi 2011).

With that said, scholars have highlighted that the projects during this period were only implemented if there was a clear economic return on the investment, as well as to maintain political stability in response to anticolonial struggles (Njoh and Akiwumi 2011). Evaluation of discourse and function of WaSH development therein thus offers useful context for identifying vestigial themes in time periods discussed hereafter.

WaSH under 20th century colonialism in SSA may be contextualized materially, geographically, and intersubjectively. First, the level of technological advancement at the time maintained a relatively homogenous material distribution. Centralized urban developments like water supply and distribution piping, sewers, and flush toilets were the dominant strategies employed (Nilsson 2016, Kithiia and Majambo 2020). Within rural areas, deep wells and distribution piping were also dug, but most development of this kind was focused on growing a colonial agricultural sector rather than addressing the health concerns of its inhabitants (Nyanchaga 2016). Native dwellers were thus largely expected to depend on whatever natural sources were available as WaSH access was not yet viewed as a significant challenge in these geographies as compared to urban ones on account of their lower population densities (Black 1998, D. Hodgson 2001a, 48-92). Importantly, this is not to say that such a belief was true; exposure to contamination accelerated along with that of the colonists' agricultural, mining, and tourist practices (Mbilinyi 2016, Yeleliere, Cobbina and Duwiejuah 2018).

Nevertheless, colonial investment in WaSH infrastructure was resultingly geographically heterogeneous. Specific places and populations received higher levels of development financing than others depending on their economic value to the metropoles (Njoh and

Akiwumi 2011, Nilsson 2016). For example, a systematic review of both French and British colonial investment portfolios from Ricart-Huguet (2021) found that finances were directed largely to port cities that facilitated easier trade with Europe and housed larger populations of colonial administrators. Both British and French administrations also reportedly favoured certain local ethnic groups that were less resistant to their occupation, rewarding land, as well as health, education, and infrastructure programs (Njoh and Akiwumi 2011, Mathur and Mulwafu 2018, D. Hodgson 2001a, 48-92). Rural areas and their inhabitants were furthermore comparatively underserviced (Nilsson 2016), laying a foundation for a rural-urban material discrepancy.

How the select benefactors were granted access to WaSH services was also notably paternalistic. Colonial administrators widely believed that the lack of 'modernity' among the indigenous populations meant they were incapable of determining their own desired outcomes and could not be trusted to act in their own best interest. Local spirituality and belief systems were widely denigrated, with those practicing 'traditional' health measures being ostracized as immaterialist, unscientific, and valueless (Yeleliere, Cobbina and Duwiejuah 2018, Fofana 2020). Programs were consequently implemented explicitly *for* locals with limited participation beyond expected financial contributions and labor, which were themselves often unreasonable (D. Hodgson 2001b, 100-139, Tilley 2016, Mathur and Mulwafu 2018). As such, though WaSH-related developments among SSA populations certainly accelerated in this period, it transpired beneath a veil of dismissiveness that actively counteracted the positive gains accrued from their material implementation.

A clear example of exclusionary colonial WaSH practice is the 1928 piping project to transport water from Lake Victoria to Kampala, Uganda. Though framed to policymakers in the British metropole as a crucially important public health project, no local consultations were completed, nor was local health targeted. Rather, the project was approved to "provide ample consumption per head to the non-African population," with opportunity to expand distribution to neighbouring local communities if sufficient finances could be levied to construct and maintain a "modern township" (Nilsson 2016). 13,500 m^3 of water (10x European demand) was subsequently piped daily to serve the "white" population while most locals, particularly in the rural outskirts, continued collecting rainwater or drawing from few constructed boreholes, where available (ibid.). Water infrastructure was thus used to demarcate material inequity between colonizers and colonized and emphasize that access to superior technology was reserved for the *modern* individual unless *earned*.

8.4.2 Post-War Transitioning (1948-1978)

With the close of World War II (WWII) came United States (US) President Truman's famed Marshall (1948) and Point IV (1949) Plans and the official beginning of the international development era (Leclerc 2007). Meanwhile, communist-socialist forms of political organization were gaining popularity among colonized populations as the struggle for global power between the US and Soviet Russia (USSR) was intensifying (Lorenzini 2019b, 33-49). Western powers were thus newly motivated to evoke an appearance of benevolence on the world's stage and infrastructure expansion and technical knowledge transfer became vehicles for doing so (Wethal 2019). Development was no longer only a question of economic, social, and humanitarian consideration, but a means of persuading alignments

Westward. Truman and his European counterparts invested heavily in research that could simultaneously lift people out of what was now called "poverty" and prove that market freedom, democracy, and liberalism were specifically superior to socialist alternatives (Lorenzini 2019c, 22-33). Post-war international development policy was moreover designed to instill the belief among colonized populations that changing to follow Western knowledge, practice, and behaviours would facilitate material gain.

The Maasai Development Program (MDP: 1949) is a clear such example. The MDP was a devised by British administrators in Tanganyika to address material deficits facing Maasai communities. Namely, water access/usage, economic sustainability, and population health were identified as key areas of interest by the colonists. Administrators therefore developed targeted programming that aimed to change local animal husbandry and water management practices to more closely resemble Western modes of production, *modernizing* the populations to *improve* local conditions. Specifically, beginning in 1949, the Tanganyikan administration invested in water infrastructure, land clearing, and cattle-specific veterinary programs, which were believed to promote livestock productivity and thus improve economic viability and social development via market forces (TNA 1950). Excited particularly by the prospect of increased water access, Maasai communities contributed £65,000 ($1.5 million in 2022 USD) and tens of thousands of cattle to develop the initiative, in addition to paying a 55-shilling tax per participant each year (the highest in Tanganyika). Communities then accessed water project financing if they demonstrated Western-style animal husbandry, contributed unpaid or underpaid labour to clearing new grazing lands, and took ownership of the water supply so as to assume responsibility for maintenance costs.

Many Maasai, however, resented the forceful nature of the program's conditional implementation and felt it was unfairly burdensome relative to the return. Participation resultingly decreased with time and the significant local investments saw very little tangible impact: by 1955, less than half of the estimated grazing acreage for cattle was usable and only 40 water projects (boreholes, handpumps) had been built across the entire Maasai Reserve, almost all of which had broken down (TNA 1953, D. Hodgson 2001b, 100-139). As explained by Cooper and Packard (1997), the MDP suffered from many of the same structural issues as other projects of the time: being top-heavy, overly planned, overly managed, and distributing insufficient material through a poor institutional infrastructure.

Yet, administrators and scholars alike argued that the observed failure was a matter of Maasai production; as Evangelou (1984) said, "traditional Maasai social structure and cultural institutions fundamentally constrain development initiatives." In other words, it was that Maasai were unable to adopt Western-prescribed behaviours that limited program success. Later critics, however, demurred that the issue rather laid with the program's formulation as a mechanism for behavioural coercion towards Western models of production rather than a means of addressing the concerns of Maasai benefactors. As put by Rigby (1992), the MDP was devised such that "the road to the slavery of capitalist wage-labor was already mapped and constituted its ultimate end." Water was not about water and health was not about health - everything was about *poverty*, and *poverty* was the antecedent of *modernity*. Indeed, the MDP is emblematic of WaSH-centered international development in SSA at this time more broadly; a process driven by systematic modernization to facilitate a transition from *underdeveloped* to *advanced* societies (Nilsson 2016). Failure to facilitate that transition was

then perceived as a product of indigenous *backwardness*, *primitivism,* and/or resistance to principles of being "anti-communist, Western-oriented, and committed to nonviolence and multiracialism" (Aminzade 2013, 31-60). It was further this recipient dissatisfaction with material outcomes being interpreted by colonists as nonmodernity which, in part, galvanized movements towards decolonization (Cooper and Packard 1997, 45-63).

As colonies transitioned to independent nations throughout the 1950's and 1960's, so too did the focus of international development begin shifting. Economic and industrial growth dominated policy and planning as new states were motivated to establish an economy in the image of their own indigenous cultures. Simultaneously, Western and Soviet powers attempted to curry favour towards their models of production and political organization (Lorenzini 2019b, 33-49). For example, most of the USSR's aid efforts focused on constructing manufacturing centres in addition to some infrastructural developments. In fact, WaSH and education programs combined only accounted for approximately 3% of all Soviet contributions too SSA until 1970 (Walters 1970, 121-148). Conversely, the United States and her allies directed aid financing towards large public works projects – dams, bridges, roads – with significant emphasis on facilitating rural agricultural development (Leclerc 2007, Wethal 2019). Both were therefore chiefly focused on building the foundations for participation in an international economy, though each were guided by individuals of diverging political and ideological leanings. Regardless, investment in WaSH access decreased throughout the 1960's and into the 1970's, with behavioural and rural health programming receiving particularly limited funding. Meanwhile, country-level debt, particularly to the Bretton-Woods' International Bank for Reconstruction and Development

(IBRD), continued to rise meteorically (Volberding 2021). Rural-urban discrepancies in health-related infrastructure that had been established in times prior were exacerbated.

On May 17, 1971, discourse then began to shift again. At the opening of the World Conference of the Society for International Development, World Bank economist Mahbub ul Haq stood before global leaders and lamented how the economy-first approach had proven inadequate and the calls for more commitments from the 1968 Pearson Report[3] were misguided. "After two decades of development trying to pick up the pieces, we simply do not know whether the problems associated with dire poverty have increased or decreased or what real impact the growth of GDP has made on them," Haq argued (Lorenzini 2019d, 142-159). The push for modernization had evidently failed to address the real hardships felt by many, and indeed perpetuated the inequities produced by colonial oppression. As was famously put by Barbara Ward in 1974, the development field had become so driven by a quixotic mission to make the *Third World* in the image of the *First*, that it failed to meet the "basic needs" of its inhabitants (Lorenzini 2019d, 142-159). 'Basic needs' then became the international development sector's guiding principle in subsequent decades, launching WaSH from the periphery into the centre of discourse.

8.4.3 Appropriate Technology and Market-Based Solutions (1978-2000)

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[3] The Pearson Report was commissioned by Canadian Prime Minister Lester B. Pearson to evaluate the World Bank's development assistance effectiveness over the previous 20 years. It also provided several recommendations for future operations, largely related to calling for wealthy nations to invest in expanding access to international markets within the post-colonial nation states and reducing focus on economic growth as a metric for evaluating economic strength. The central thesis was thus that economic strength was more related to participation global commerce than year-on-year changes in Gross National Product (GNP) (Macekura 2019).

The late 1970's ushered in a new major shift in WaSH implementation policy and practice towards rural development (Black 1998, Bakre and Dorasamy 2017). The 'basic needs' discourse further led The United Nations General Assembly (UNGA) to launch the International Drinking Water Supply and Sanitation Decade in 1980 (Black 1998). This new wave of interest in WaSH was, however, emerging concurrently with a changing political landscape. That is, British Prime Minister Margaret Thatcher and US President Ronald Reagan's economic philosophies were growing in popularity and motivating aid institutions to transition to neoliberal models, replacing import substitution strategies with market-based solutions (Parfitt and Riley 2011).

Structural Adjustment Programs (SAPs) then began in the early 1980's, which added new debt to global South recipients while forcing governments to divest from public services (among other things). Increasing WaSH access thereafter thus became a challenge for mostly non-state actors (Harrison 2010): state-directed infrastructure investment dropped by 40% and privatization/NGO-*ization* of WaSH services accelerated (Spronk 2010, AfDB 2018, Mitlin and Walnycki 2020). Consequently, a significant portion of the WaSH sector was profit driven, which in turn exacerbated the marginalization of lower-income and rural individuals. Namely, industrial-scale WaSH service provision was oriented largely towards higher-income earners in urban areas who could sustain private revenues (Spronk 2010). Sparse densities and limited materials in rural communities thus meant private investment was unprofitable and not worthwhile, even though sectoral leaders were emphasizing the need to focus efforts therein (James 1980).

A different strategy was required, leading to the rise of *Appropriate Technologies* (AT) (Black 1998). AT, a rebranding of E.F. Schumacher's 'intermediate technology' (James 1980), is defined as decentralized and low-cost technologies carefully chosen to address the specific conditions of the communities within which they're implemented (Murphy, McBean and Farahbakhsh 2009). They are necessarily small in scale and less expensive than traditional centralized infrastructure (C. P. Sianipar, et al. 2013). And due to that scale, they can be easily incorporated into market- or charity-based proliferation models (Lockwood, et al. 2010). Starting in the late 1970's and into the early 1990's, technologies like pit latrines, handwashing stations, shallow wells, handpumps, and point-of-use water treatment solutions (POUWTS) gained significant traction across the global WaSH sector, thought to be the long-sought solutions to overcoming the rural-urban divide. Specifically, they were technologies that were thought to simultaneously address material need while alleviating poverty, promoting employment, and redistributing incomes (James 1980). As such, not only did AT fit within the broader narrative of meeting WaSH needs in rural areas quickly and at a low cost, but they matched the existing political-economic principle that market-based, non-governmental solutions could more efficiently address social, economic, and environmental challenges than public investment (Murphy, McBean and Farahbakhsh 2009, Harrison 2010, Hazeltine 2003).

Achieving such imagined benefits, however, proved excessively challenging. Even with the careful considerations taken for technological decision-making, programs were widely hampered by technical insufficiencies and waning public engagement. One such example is the 1984 DANIDA Drinking Water Project, in which Danish private firms, Kamphil and

Dangroup, installed 150 boreholes with India Mark II handpumps over the following four years across southern Mali. Funded by Danish and Malian government agencies, the intervention aimed to provide 40 L/person/day to 200 households across various villages in Sikasso and Kadiolo districts, as well as provide health and financial education to facilitate technological usage and promote income-generating activities. Each borehole further required village representatives to enter a contract that committed to funding its construction, monitoring, and maintenance, making this a particularly participatory program for the time. However, the 1988 review found 64% of villagers felt the produced water was insufficient for their needs in terms of both quality and quantity, which was later correlated to nearly 60% of boreholes being incorrectly dug. Further, 60% of villages did not pay for needed repairs and 86% of program participants continued to drink water from pre-existing sources rather than those implemented through the initiative (Vaa 1993). The shift in perspective from industrial to lower cost, small-scale *appropriate* technologies evidently failed to yield imagined superior outcomes therein. Similarly, the provision of education did not materialize as substantially changed WaSH behaviours.

The 1988 DANIDA review furthermore recommended improved community engagement and more behaviour-targeting education programs, as well as clearer knowledge communication regarding the economic value of the initiative. These notions were later echoed by the international community as principles of 'community participation' and 'economic valuation of water' were codified into international agreements at the 1992

Dublin[4] and Rio de Janeiro[5] Conferences and 1994 Cairo[6] Conference (Harmacioglu 2017). These agreements further aimed to emphasize the need to match technical solutions to socioeconomic conditions, as well as facilitate a sense of ownership to promote AT usage and maintenance by integrating WaSH and the environment into economic development planning. Many intended benefactors, however, remained (and remain) unmotivated to invest in WaSH solutions due primarily to their unaffordability or perceived lack of benefit (Madu 1990, Burt, et al. 2017, Albert, Luoto and Levine 2010). NGOs thus managed much of AT distribution throughout the 1990s, however due to inherent challenges with balancing scalability and financial/institutional constraints, proliferation was widely selective, disjointed, and geographically varied (Black 1998, Lockwood, et al. 2010).

The close of the millennium then evoked an energy for change. While AT remained a key part of the WaSH strategy in rural communities, practitioners and scholars recognized that it could not simply be provided or purchased; recipients had to be trained regarding why it was worth their time and financial resources to own, use, and maintain such technologies (Harmacioglu 2017).

8.4.4 The Millennium and Sustainable Development Goals (2000-Present)

The creation of the Millennium Development Goals (MDGs) in 2000 was among the most celebrated international achievements to date, setting a novel basis for how development

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[4] International Conference on Water and Environment, Dublin, 1992, ICWE
[5] UN Conference on Environment and Development (UNCED Earth Summit), Rio de Janeiro, 1992
[6] UN International Conference on Population and Development, Cairo, 1994

would proceed thereafter. And with Goal 7C, to "halve, by 2015, the proportion of the population without sustainable access to safe drinking water and basic sanitation" from 1990 levels, WaSH became central to international progress. This importance was also emphasized at the Rio +10 Conference[7] in 2002, as well as with the creation of 'The International Decade for Action: *Water for Life*' in 2005-2015 (Harmacioglu 2017) and the establishment of water and sanitation access as a human right in 2010 (Bakker 2012, 19-44). Behaviour change, however, was now an explicit development goal: practitioners were urged to (1) "promote safe hygiene practices," (2) "promote education and outreach focused on children as agents of behaviour change," and (3) "promote affordable and socially and culturally acceptable technologies and practices" (UN 2002, Harmacioglu 2017). The MDG era offered a new lens on WaSH development at the individual level, where roadblocks to progress, particularly in rural areas, were newly viewed as behavioural challenges with a critical technical and technological component rather than its inverse as before. Put another way, the governing theme of the WaSH sector borne out of the MDG discourse, which still persists today, was that practitioners must assist individuals to change hygiene and water/sanitation technology usage behaviours to achieve meaningful progress in associated access. WBC research then became the study of how to do so (Dreibelbis, et al. 2013, UN 2015, UNICEF 2016).

A clear example of an MDG-era program that embodied this change in WaSH perspective was the Millennium Water and Sanitation Program (*Programme d'Eau Potable et*

[7] World Summit on Sustainable Development, Rio+10, Johannesburg, 2002

d'Assainissement du Millénaire au Sénégal; PEPAM) in Senegal. Implemented by Research Triangle Institute (RTI) and funded by the United States Agency for International Development (USAID), PEPAM studied the impacts of three different AT delivery models related to the implementation of handpumps, POUWTS (e.g., Aquatab), pit latrines, and handwashing stations serving 10,000+ households between 2009 and 2014. However, differing from earlier AT programs, PEPAM involved significant WaSH education, particularly in terms of hygiene and sanitation behaviour, as well as AT use, repair, and maintenance (USAID 2019). Specifically, workshops followed the WHO's participatory hygiene and sanitation transformation (PHAST) and self-esteem, associative strengths, resourcefulness, action planning, and responsibility (SARAR) approach, which is among the most popular WBC programs for facilitating behaviour change among both adults and children.

The program was resultingly quite successful in in some areas, yet unsuccessful in others. For instance, 84% of participants reported satisfaction with the quality and quantity of water produced by the handpumps. However, this level of satisfaction was closely related to the type of handpump and borehole; deep wells with the higher flowrates and handpumps with less reliability issues (e.g., maintenance requirements) were those most favoured. Only 27% of participants were satisfied with the least expensive handpumps, Erobon rope pumps, and factors such as reliability and the physical demand to retrieve water were leading causes of disuse. Additionally, only 27% of users treated their water with a POUWTS, the majority of whom used the simple, accessible, and rather ineffective technique of sieving through a cloth (USAID 2019).

Impacts of the sanitation program also yielded similar results. That is, where latrines were installed within walking distance of the home, usage rates were nearly 90% across all villages and implementation strategies. Rates of latrine construction and maintenance were further superior in communities with construction subsidy programs. In fact, the majority of interviewees without a latrine reported unavailable funds as the primary barrier to their construction. Further, of those reporting practicing open defecation, >70% were because latrines were not accessible or usable while 6% were due to habit continuation. Finally, the hygiene intervention proved the least successful, with only 6% of households reporting the construction and/or maintenance of tippy-tap handwashing stations (low-cost structures that use a pedal to *tip* water over for handwashing). No tippy taps were observed as in use at the project's end. And despite a marked increase in retained knowledge related to handwashing protocols among education program recipients, associated behaviours did not differ from their non-participant neighbours.

Participant behaviours were furthermore consistently referenced as key drivers of programmatic challenges. For instance, the report reads, "it could be possible that the way different implementers carried out their approaches impacted outcomes (e.g., one may have been better at behavior change communication)," when discussing sanitation outcomes. Reviewers therefore posited that of the diverse factors that may lead to latrine disuse, a lack of motivation was chief among them. Similarly, the report later states, "PEPAM's behavior change appeared to be insufficient to establish handwashing habits and norms" (USAID 2019). Meanwhile, an alternative reading of the data suggests technological inadequacy was perhaps more relevant than considered. Laborious handpumps were commonly unused; time-

demanding POUWTS were never adopted; maintenance-demanding pit latrines were abandoned; notoriously precarious tippy-taps were left unfixed. Indeed, solutions that created additional physical, time, and knowledge burdens on individuals were those which resulted in the lowest rates of usage and satisfaction (USAID 2019).

With that said, PEPAM, like diverse other programs borne of the MDG discourse, did produce powerful improvements in the lives of many of the most disenfranchised individuals. By 2010, the water portion of MDG 7C was achieved (UN 2015). Yet, as shown in Figure 8.1, the disproportionate marginalization of those in rural SSA persists, with the rate of change remaining insufficient for reaching access equity. For instance, in 2015, where 10% of all individuals lacked basic drinking water and 22% lacked basic sanitation across the world, 51% and 78% lacked those same services in rural SSA, respectively. As such, despite achieving important progress since 2000, the MDG era notably failed to substantially reduce WaSH-related inequalities across the geographic and economic landscape (UN 2015).

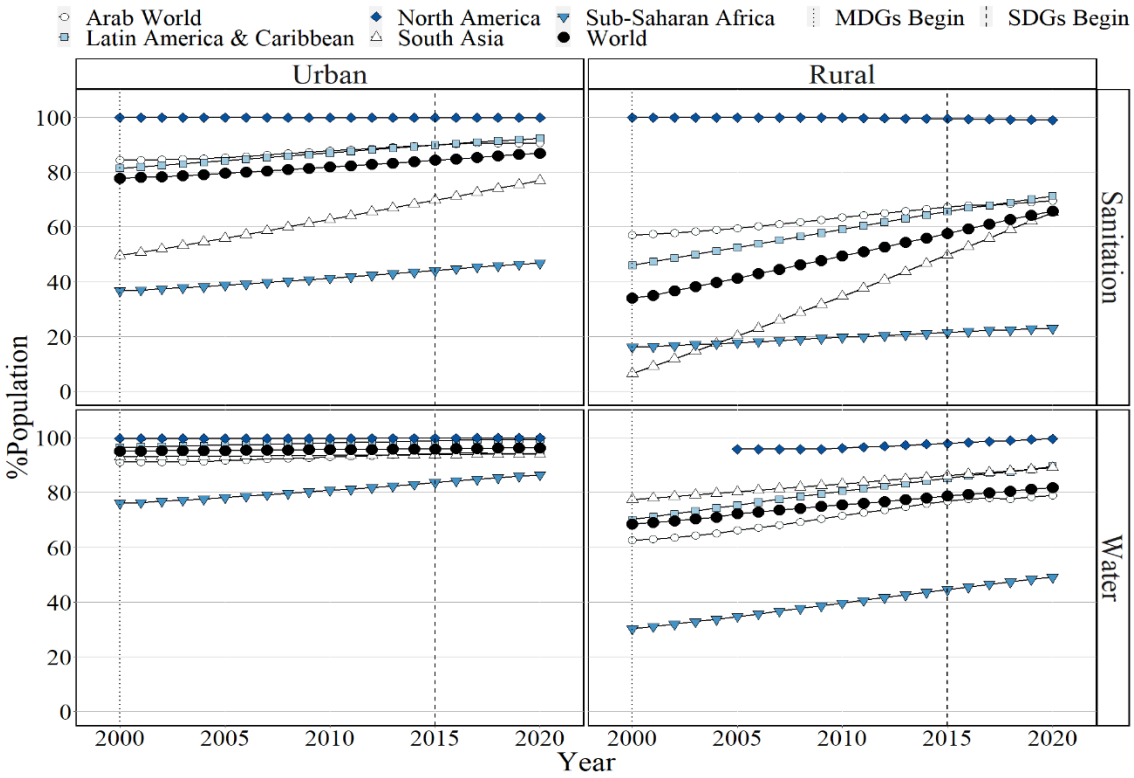

Figure 8.1 Percentage of Population with access to basic drinking water and sanitation services, 2000-2020 (Data: World Bank 2021)

The transition to the Sustainable Development Goals (SDGs) in 2015 again took a different energy than its predecessor, creating a new framework for WaSH development. Water, sanitation, and hygiene each received unique targets with more refined definitions, and the metric for evaluating access became 'safely managed services' rather than 'improved water' and 'basic sanitation.' WaSH programs were also newly encouraged to incorporate 'local ownership and administration' in addition to 'participation' as per the 1992 Rio Conference (UN 2015), moving away from the provisional model of development. The SDG language thus critically refocused WaSH sector objectives to look beyond Ward's 'basic needs' towards establishing conditions in every geography – rural, urban, high-income, low-income

– as requiring equal levels of service with equal levels of programmatic control and involvement.

Yet, discursive accomplishments aside, achieving such laudable goals appears highly unlikely: current trends project the world to miss the drinking water target by 6% and sanitation target by 11%. Within rural communities in SSA, the margin increases to 45% and 75% for drinking water and sanitation, respectively (World Bank 2021). Progress in language is thus evidently failing to translate to the required progress in practice.

8.5 Discussion: How Policy Influences Practice

The relevance of individual behaviours in the transmission of biological disease is uncontentious, yet WaSH practitioners continue to debate whether programs should be targeted towards promoting behaviours that reduce risk of transmission or addressing the conditions from which such risk evolves. In this essay, I detail historical discourse, policy, and programming within the WaSH sector and highlight key factors contributing to how inequal risk exposures and the means employed to address them have emerged over time. I further demonstrate that both psychological and power-relational characteristics of development as an emergent phenomenon of colonial occupation, as well as time-specific political-economic pressures have shaped the persistence of material inequality and the reliance on WBC as a mechanism for overcoming it.

Namely, colonial underinvestment in rural WaSH, as well as geographic sparsity, produced conditions where less material resources were available in these geographies per individual than their urban counterparts. Meanwhile, colonial denigration of local capacities instilled a

belief among native populations that local modes of production were inferior to those of Europeans. Colonists thus manufactured an imagined legitimacy of white supremacy and defined development as prescriptive process towards Western model replication via expertise from external actors. During the post-war infrastructure boom, Western development policy and economic superiority then built upon this power inequity to plan and implement infrastructure projects that could guide economic growth. Consequently, despite evident local interest in rural WaSH (as exemplified by the MDP), its direct economic contribution was considered limited and thus political interest and associated implementation were limited by extension. The economic policies during colonial occupation furthermore created a North-South and rural-urban material disparity that facilitated sustained epistemic hegemony when the era of *development* began, as well as in the decades thereafter.

A similar relationship between policy and practice is observed within the rise of 'basic needs' discourse. International institutions maintained Western economic thought leadership and produced a new paradigm through SAP loans and neoliberalism more broadly. The policy attachments to these loans then ensured WaSH was addressed predominantly by foreign private and non-profit sector actors. Again, though its language was markedly different than its predecessors, this third wave of international development policy held decision-making power within wealthy nations in terms of both national policy development and implementation. Responsively, WaSH programs were largely disjointed and uncoordinated at the community-to-community level and practitioners were forced to rely upon interventions determined by each organization's operating budget. Where infrastructure investment remained unprofitable or infeasible, low cost and small-scale AT appeared a

natural choice. Yet, as exemplified by DANIDA, AT usage and maintenance was difficult to facilitate and associated programmatic impact was insufficient.

The subsequent turn towards WBC programming can further be viewed as per these same processes. Namely, while funding for WaSH increased substantially with the MDGs, that amount has remained inadequate to meet service demand (Alaerts 2019). Similarly, interest in community participation and involvement in programming flourished, yet local programmatic ownership remains exiguous; WaSH services are still predominantly provided by private and non-profit actors beholden to meager budgets, donor interests, and profit motives (Martin, et al. 2018, Spronk 2010, Lockwood, et al. 2010). Summarily, neoliberalism and the subsequent means of WaSH implementation have persisted (Harrison 2010). As such, with substantial progress still required to achieve WaSH equity but limited new material resources being allocated thusly (OECD; UNDESA 2020), practitioners and academics have aimed to improve upon existing solutions through WBC. Yet, as evidenced by PEPAM, promoting WBC as a solution to AT inadequacy invokes what Richardson (2019) defines as *coloniality*,

> the matrix of power relations that persistently manifests transnationally and intersubjectively despite a former colony's achievement of nationhood [where] hierarchical orders imposed by European colonialism have transcended 'decolonization' and continue to oppress [through the maintenance of] bourgeois empiricist models of disease causation, which serve protected affluence by uncritically reifying inequitable social relations […] and making them appear commonsensical.

Put another way, WBC promotion necessarily presupposes that the knowledge already possessed by target populations is inadequate to realize safe WaSH access and knowledge communication regarding WaSH behaviours is required to address a gap in understanding. This notion thus echoes the sentiments proposed by colonial administrators – as observed in response to MDP failures – and similarly commits "hermeneutic injustice" (Pohlhaus 2012) by interpreting past programmatic shortcomings as a function of the individual and the mechanisms employed to change them. AT's *appropriateness* is assumed self-evident. Just as DANIDA's failures were understood as resultant of participants *choosing* to continue using their traditional water sources rather than fix those built during the program; just as PEPAM's failures were understood as resultant of individuals *choosing* to not repair broken pit latrines or tippy taps and *choosing* to not use POUWTS. They are interpretations couched in a perspective that programmatic recipients' modes of production are fundamental determinants of programmatic outcomes, normalizing inequality in turn. Failure is then further conceived as a function of insufficient WBC; as the PEPAM report concludes, "the [tippy tap] handwashing stations PEPAM promoted no longer exist and replacement has been limited. With less than half of all observed [households] possessing any materials or facilities to wash hands and in spite of self-assertions regarding handwashing practices, the behavior change strategy did not appear sufficient to change handwashing behavior long-term" (USAID 2019). Meanwhile, the report omits that, perhaps, individuals were simply not motivated by low-quality and labor-intensive solutions (i.e., AT). Perhaps investing their limited resources in repairing a solution that was difficult to use and certain to break again was not worthwhile. Perhaps the expectation that individuals should bear the construction

and maintenance costs of WaSH solutions is itself unsustainable. In other words, if access to WaSH service is indeed a human right, then placing the weight of its realization upon those without it is paradoxical; a burden is added to overcome another (Ray and Smith 2021). And as exemplified by the widespread usage of high quality and subsidized water points and latrines under PEPAM, as well as the following disuse and disrepair after their failure, doing so is demonstrably ineffective. To meet the ideals put forth by the SDGs, the WaSH sector must reform.

8.6 Recommendations

Ray and Smith (2021) recently stated "public health researchers have had an oversimplified understanding of poverty; we have [ignored] uncertainties, stresses from constant scarcity, and attendant fears," responsively recommending that "rather than improved versions of household-scale delivery models, transformative investments in safe water for all […] require utility-scale service models." The authors are thus promoting a radical departure from the sector's methodological orthodoxy towards a model that is nearly ubiquitous in the global North (World Bank 2021): that safe water and sanitation access be provided and maintained by a trusted authority. In rural SSA, however, an expectation that the individual bear the responsibility of achieving their own safe WaSH access is normalized: one must assume the burden of purchasing a WaSH solution, the burden of maintaining the solution, and the burden of learning new skills and information to ensure that maintenance is performed effectively. Simultaneously, the solutions themselves are commonly ones which practitioners from the global North would not accept at home.

As has been demonstrated through this writing, these differences in convention between rural SSA and higher-income geographies are not coincidental but the result of persistent social and economic coloniality. The calls put forth by Ray and Smith (2021) are therefore of imminent importance for the WaSH sector to evolve beyond well-intentioned exponents of colonial violence to stewards of human rights who work to serve all benefactors equally and without discrimination. Significant methodological and economic reform are further required, some recommendations on which are provided below.

8.6.1 WaSH as Service

Of the various factors challenging sustainable WaSH access, ease of technology use consistently ranks among the most influential (Ray and Smith 2021, Burt, et al. 2017). Individuals seldom choose to increase their daily burdens unless the return on their time and labour investment is evident and remains evident over time (Middleton, Anton and Perri 2013). And while diarrheal illness has significant mortality and morbidity implications (GBD Risk Factor Collaborators 2020), acknowledging its absence is often challenging. That is, one tends to observe the impact of changing behaviours – e.g., to use AT – in contrast to recent recall of diarrheal presence (Martin, et al. 2018). Otherwise, resorting to the practice that requires the least effort – e.g., drinking water from its source – may be expected (Ginja, Gallagher and Keenan 2021). WBC programs attempt to correct this situation by providing information and resources to make the benefits of initiatives more obvious to users, *nudging* them towards specific WaSH practices. However, when viewed within an historical context, this strategy appears more of an approach to overcoming programmatic inadequacy than WaSH inequity. Rather than attempt to change individuals, practitioners must instead expand

WaSH programs to ensure the easiest option available to the individual is that which provides safe access. Specifically, it is recommended that the WaSH sector:

- **Increase centralized[8] WaSH utilities in rural areas.** In the long-term, rural areas of every country must equally have easily accessible and high-quality WaSH services. Despite opposition to centralization due to practical limitations, no other solution reaches people close to their homes with treated water, safe sanitation, and hygiene services as effectively or efficiently (Milman, Kumpel and Lane 2021). As such, infrastructure construction must drastically expand, and future research must investigate models where centralized and semi-centralized water and wastewater treatment and distribution utilities may be delivered to rural residents at a sustainable cost.
- **Provision of AT must be accompanied by robust monitoring, maintenance, and user-support protocols**. While addressing infrastructural inequities in rural areas is inevitably time-intensive, AT remains useful in addressing immediate-term needs. The WaSH sector should, however, move away from models for facilitating behaviour change to ones where the factors leading to disuse (e.g., breakage, time burdens, financial burdens) are directly managed by trained personnel. AT should therefore be maintained and monitored regularly by water providers who are locally governed and operated. Water providers should also directly address technology-

[8] Centralization refers to the water and wastewater treatment and distribution systems that operate from a single facility or network of facilities.

related challenges on an individualized level via demand-driven response mechanisms. Alternatively, water providers should maintain complete control of AT and operate quasi-centralized facilities. In either case, the primary objective must be to alleviate the burdens of care from the individual. Further research into governance, monitoring, support, and funding structures of such programming is required.

8.6.2 A Brief Note on Economic Reform

As has been detailed throughout this writing, the WaSH sector is constrained by political and economic pressures that have limited the breadth and types of solutions implemented by both public and private institutions. WaSH access inequities do not exist in a vacuum and cannot be meaningfully overcome without too confronting the unjust economic conditions from which they manifest. Providing specific recommendations on how to reform such a system is, however, beyond the scope of this review. Rather, further exploration of heterodox economic models for facilitating socially and environmentally conscious development is suggested. For instance, post-Keynesian and Ecological Economics movements (see (Hudson 2010, Berr 2015)), as well as movements towards international debt forgiveness (see (Thomas 2000, Ndikumana 2004, Harrison 2010)) are considered promising avenues worthy of investigation.

8.7 Conclusions

This review detailed WaSH discursive history from 1918 to 2021, with specific focus on rural SSA, to elucidate the respective roles of individual behaviours and policy/programming decisions on the manifestation of water-related inequality. More specifically, this work

intends to illuminate an ongoing debate within the sector regarding driving factors of disease transmission and optimum solutions for mitigating the associated risks. Key findings are:

- Colonial development policies concentrated WaSH infrastructure in urban areas and facilitated the material discrepancy at the root of current rural-urban divides.
- Modernization theory extended through late colonialism into post-colonialism and informed development policies at national and international levels such that economic growth was the primary motivation.
- Attention to rural WaSH rose with neoliberal economics, which fueled the implementation of lower-cost technological solutions (AT) by largely private and non-profit actors.
- Unsustained usage and maintenance of AT led to the methodological turn towards WBC.
- WBC perpetuates themes from modernization discourse by targeting user behaviour to overcome technological disuse without concurrently addressing the unequal technological and environmental conditions from which such behaviours manifest.

Building from historical lessons, this work furthermore recommends that (1) programs adopt a service-oriented approach to WaSH access that works to alleviate the burdens of its recipients, and (2) political and economic leaders adapt to existing circumstances and invest in human flourishment over the continual expansion of wealth among a concentrated few.

8.8 Acknowledgements

I want to thank Dr. Onita Basu and the Basu Research Group, as well as my brother, Sam, for their ongoing support. I also wish to dedicate this paper to all members of the Longido community in Tanzania who have provided me with the friendships and life learnings that motivated this work.

Chapter 9: Summary, Reflections, and Unsolicited Ruminations

This dissertation details a multidisciplinary approach to addressing challenges with safe drinking water access in remote and resource-restricted contexts. Throughout its production, I attempted to bridge exploration of technical methods for innovating a point of use water treatment solution – the ceramic water filter – with reflexive and participatory investigation of key drivers impacting technological usage and associated child and maternal health.

Chapter 4 demonstrated that combining silver and zinc oxide nanoparticles under batch conditions, as well as when impregnated into CWFs, can synergistically disinfect *E. coli* across varying water qualities. Namely, combined Ag-ZnO mixtures consistently yielded superior disinfection outcomes than either MNP species in isolation. This study therefore highlighted that if ZnO is appropriately applied in conjunction with Ag, CWF cost may be reduced substantially while maintaining or increasing disinfection efficacy during storage. What has remained the dominant paradigm for more than 40 years – that CWFs be impregnated solely with AgNPs – was thus newly challenged in a meaningful and optimistic way. Additionally, the outcomes illustrated that DO particularly influenced nanoparticle disinfection efficacy. That is, bactericide worsened by more than a factor when DO was lowered to just above hypoxic conditions. I therefore showed how profoundly influential water chemistry is on MNP disinfection impacts and questioned the CWF treatment capacity robustness upon which researchers and practitioners often depend. Put another way, the work highlighted a need to better understand how CWF water treatment may change according to factors that influence water quality like seasonality, geography, and contamination source.

The research described in Chapter 5 intended to address remaining uncertainty regarding the translation between impregnation and elution with co-fired ceramic disk- and pot-filters by quantifying the concentration of released metals and isolating their relevance to disinfection during storage. While several studies have enumerated Ag elution from CWFs (particularly those painted), this was among the firsts to do the same with an alternative MNP, and ZnO in particular. It further contributed to the very small body of work that has evaluated co-fired CWFs and was the first known study to relate elution results with associated bactericide. The outcomes moreover confirmed the opportunity for reducing filter cost through ZnO supplementation. Perhaps more interestingly, though, were the observed presence of background Zn concentrations within the clay matrices and notable differences between Ag-Zn concentration and disinfection found during disk- and batch-scale experiments. These results suggest clay chemistry may play a more profound role in CWF water treatment than previously considered. What began as a study to isolate and clarify the roles of impregnated AgNPs and ZnO on disinfection during storage became a first clear demonstration of CWF effluent matrix complexity and its associated relationship with bacterial treatment. The work may therefore afford opportunity to expand the theoretical horizons from which CWF function and manufacture are investigated.

Chapter 6 diverged somewhat from previous chapters by focusing on physical filtration rather than MNP disinfection. Specifically, it is the first study known to use a nested multiple regression analysis approach to relate input materials to performance outcomes, and further, the first to use machine learning with fuzzy logic to predict those outcomes. This work can improve quality assurance (QA) and quality control (QC) methods by offering a novel

framework with which flowrate, bacteria removal, and material strength are predicted. That is, if easily and inexpensively measured parameters like porosity and density are utilized, CWF performance may be more accurately estimated, as well as more easily compared across manufacturing settings. The development of these models may thus be an important first step towards methodological standardization and consistent production of high-quality filters across geographies.

In summary, this technical work has established existing challenges within the research field, proposed novel solutions for overcoming some of them, and demonstrated the value that these solutions offer. From the critical literature review publication (Chapter 2) that summarizes data from across over 100 sources and adds clarity to the role of MNPs in CWF processes to the two studies on combined AgNP/ZnO disinfection within batch and impregnated filter phases (Chapter 4 & Chapter 5) and the investigation of a CWF performance model with machine learning (Chapter 6): each of these works has advanced CWF research by demonstrating somewhat novel phenomena and highlighting important pathways for future exploration. As such, I do indeed believe I have contributed to progress *towards an improved ceramic water filter paradigm* in this capacity.

Meanwhile, the insights from the social scientific research presented in Chapter 7 and Appendix B are also highly informative to modern WaSH practice and CWF implementation programming generally. That is, our case study showed that participatory and locally controlled decision-making, as well as long-term, culturally relevant, and repeated knowledge communication are critical programmatic features for promoting CWF longevity. Data gathered among members of the Hospital Clinic Group in Appendix B further showed

that singular information and filter provision was broadly insufficient to promote technological usage, maintenance, and protection. Additionally, the process of conducting the research and community consultations revealed that factors like the sociodemographic characteristics of knowledge communicators, the existence and structure of indefinite participant support mechanisms, and programmatic agility to adjust to participant needs were important for achieving a positive reception to the intervention. The study therefore illustrated important insights regarding methods for meaningfully engaging with community members and developing a locally driven intervention that facilitates long-term behaviour change and behaviour change maintenance; it was indeed a feat in addressing locally identified needs through locally identified solutions.

Separately, the overarching project objective to promote filter adoption across the Longido population, as per the request and interest of community leaders and consultees, was also met. For instance, when I started my degree, not a single person in Longido was willing to pay for a filter; nearly 20 have done so in the past two years alone. When I first began meeting with nurses and doctors at the Longido District Health Centre, they didn't want to promote the filters because they didn't know enough about them; a filter now sits in every ward and staff openly recommend their purchase to patients. It was the process of taking time to slowly work with community members and leaders across educational, political, public health, and non-profit sectors that made these important gains possible. I remain steadfast in my conviction that similar slow and highly engaged approaches should be taken in the development of any such intervention.

With that said, the observed fluctuations in health with WaSH knowledge communication, as well as correlations between program attendance, filter breakage, and information retention, lead me to question the value of household-scale water treatment more broadly. That is, these results indicate that long-term and repeated education is critical to facilitating technological adoption and associated behaviour change. Or in other words, CWF sustainability is predicated on an individual's willingness and ability to assume additional time and effort burdens. But why should one be expected to provide this level of commitment to access their fundamental human rights? Is it not somewhat unreasonable to suggest that resource-restricted and remote individuals be regularly available to receive information and training so they too may access services which I enjoy with minimal effort? Or as I discuss in detail throughout Chapter 8, why should households generally assume the additional responsibilities and stresses related to using, maintaining, and protecting a technology that is, frankly, likely to break regardless of their protective efforts?

My confliction is furthermore not with whether we achieved our objectives but whether the objectives themselves were the right ones to truly advance *towards an improved CWF paradigm*. Namely, the notion that behaviour change had to be promoted to sustain positive health outcomes is itself paradoxical. Learning a new skill and adopting a new behaviour necessarily demands time, labour, and mental capacity. To rely on such a process to promote safe WaSH access is thus creating a burden to alleviate another. As such, if one must actively engage in activities to change a recipients' habits to make a solution sustainable, perhaps it is rather the solution which is unsustainable. Perhaps it is a false assumption that household-scale water treatment will advance progress towards safe drinking water access in the

immediate term. Perhaps the central problematic is not the means through which this technology is presented to the household but that it is inappropriate to present such a technology to the household at all. Moreover, I am considerate of an embedded coloniality guiding the provision of seemingly inequal and/or inadequate WaSH solutions to remote, resource-restricted communities *because* they are remote and resource-restricted. As I consider throughout Chapter 8, does household scale water treatment address the needs of its users as intended, or does it, as Richardson (2019) says, "reify inequitable social relations and make them appear commonsensical"?

In conclusion, on whether I advanced progress towards an improved ceramic water filter paradigm, I guess the answer is yes and maybe. I feel highly encouraged by the technical research that we have developed surrounding MNP impregnation and performance characterization. There are further actionable outcomes from this work that could be implemented at CWF factories, as well as various avenues worthy of continued research exploration. Meanwhile, the way in which my field research engaged with the community and facilitated program development led by local stakeholders and guided by locally identified needs is worthy of celebration. It has been a collaborative effort among all who have been involved. I am overwhelmingly grateful to have had the opportunity to participate and deeply proud of the contributions we have made to the Longido community and WaSH sector more broadly. All WaSH programming would benefit from the level and type of community engagement that we were lucky to have enjoyed. But does an improved CWF paradigm include household-scale delivery and behaviour change programming? Sometimes, perhaps. But certainly not always, and certainly not as a permanent solution.

Chapter 10: Conclusions and Recommendations

10.1 Conclusions and Significant Contributions to the Field

The objectives of this research were to (1) evaluate ZnO as a replacement or supplement to Ag to reduce the cost of co-fired CWFs, (2) identify optimal CWF material parameters that maximize flowrate while maintaining adequate bacteria removal and strength characteristics, and (3) investigate key drivers for sustained CWF usage and associated positive health outcomes using a participatory implementation and WaSH knowledge communication model. Chapters 4-7 further addressed each of these goals through systematic evaluations of natural and social scientific phenomena, while Chapter 8 explores the historical epistemological foundations from which this research emerged.

Significant natural science/engineering outcomes and contributions include:

- While ZnO provides insufficient bactericidal capacity in isolation, its combination with Ag offers significant opportunity to reduce Ag quantity within CWFs. CWF manufacturers could therefore benefit from significant cost reductions by supplementing Ag with ZnO. Best impacts may be observed when combining MNPs in 67% Ag, 33% ZnO ratios, and when ZnO is dissolved before impregnation. Effluent concentrations of 10 ppb Ag and 160 ppb Zn may further result in the safest water quality over 72 hours of storage. However, more research is required to refine these ratios to determine an optimum quantity of each (see Section 10.3).

- Water quality, and dissolved oxygen specifically, critically impacts MNP disinfection efficacy. If challenged by water with high concentrations of nitrogen, faecal matter, or other oxygen-depleting compounds, CWF disinfection achieved by filtration may not be maintained during storage, increasing risk of unsafe water consumption among users.
- Background zinc concentrations were observed for the first time in this work (to my knowledge), suggesting clay elemental composition may play a more significant role in CWF water treatment efficacy than previously considered. Namely, CWF effluent matrices may include other metallic species commonly found in clays (e.g., Al_2O_3, MgO) that could contribute to disinfection during storage.
- CWF performance modelling is superior with generalized material parameters than if using clay-sawdust ratios, FM sizes, or flowrate. Given the data at hand and associated model outputs, exploration of CWFs with the following ranges of material parameters would be most beneficial:
    - Porosity: 39.2-47.6%
    - Dry Density: 1.17-1.34 g/cm^3
    - Intrinsic Permeability: 3.61 x 10^{-9} - 16.72 x 10^{-9} cm^2
    - Clay-Sawdust Ratio: 42.2% - 47.3% Clay (by volume)
    - Nominal Sawdust Particle Size: <0.595 μm

Critical social scientific outcomes include:

- While child and maternal diarrheal illness remains a critical challenge across the Longido District of Tanzania, CWF implementation significantly improved water-related health. Diarrhea, however, fluctuated with the provision and pausing of participatory and multi-week knowledge communication programming. Cyclical, repeated, and long-term WaSH education was thus shown to be critical in promoting WaSH information retention and associated positive health outcomes.
- Water-related health outcomes improved among participants with increased time and knowledge communication, but the program was challenged to yield the same benefits among their children. These observations therefore highlight that behaviour-targeted education and water treatment technology implementation alone are insufficient to eliminate risk of diarrheal illness across a community landscape.
- While programming to facilitate behaviour change and behaviour change maintenance are crucial to WaSH intervention sustainability and POUWTS sustainability specifically, the associated burden of change thrust upon intended benefactors raises ethical concerns which must be reconciled across the WaSH sector.

10.2 Limitations

While this research intended to direct progress towards an improved paradigm of CWF production and implementation, it is certainly not exhaustive. For example, only Ag and Zn nanoparticles were investigated within this research despite the dozens of alternative MNPs which may also enhance disinfection (e.g., copper, titanium, magnesium, gold, etc.). Similarly, within these two MNP species, only one type of each was evaluated. Investigation of different shapes, sizes, functional group arrangements, structures, charges, surface

modifications, stabilization agents, and other characteristics was outside of the scope of this work. Further investigation of these parameters is recommended.

Another study limitation was that of the input material parameter investigation. Namely, materials and manufacturing methods evaluated within this work were limited to those currently used by our industrial partners in Ngulelo, Arusha, Tanzania. CWFs may be fabricated using different clay types, different FM sizes, different firing techniques, and different mixing techniques than those used herein. Evaluation of these parameters was, however considered outside of the scope of this research.

Finally, field research was significantly limited by a diversity of factors, COVID-19 being chief among them. For instance, plans for additional data collection via qualitative interviews, water quality sampling, and climatic data collection were foregone due to travel restrictions. Assessment of factors such as social cohesion, remoteness, CWF field performance, and weather were resultingly deemed outside of the study scope. Finally, there are certainly additional factors (i.e., unknown unknowns) that were not assessed or considered due to my inability to be present within the field and learn about them. The scope of the intervention was thus necessarily limited by extension. Similarly, data collection for the third intervention group was delayed due to COVID-19 gathering restrictions and thus its presentation within a publication format was not possible. Preliminary evaluation of these outcomes is, however, presented in Appendix B.

10.3 Recommendations

Several results and observations throughout this research have clearly illustrated a need for further work on improving the design and methods of implementing ceramic water filters. From Chapters 4 and 5, key questions remain regarding how to impregnate CWFs with Ag and ZnO such that the appropriate concentration of each is eluted and the desired effects are achieved. Investigation of varying Ag and ZnO impregnation concentrations is thus required. Additionally, as this research is the first known to evaluate combined Ag and Zn impregnation within CWFs, more studies are needed to improve the knowledge related to how such filters perform in general. Exploration of Ag/Zn elution and associated disinfection under diverse water qualities is particularly recommended. Separately, improved characterization of CWF effluent matrices is also needed. That is, the observed release of unimpregnated Zn from the CWFs demonstrated that clay elemental composition contributes to the physicochemical properties of the filtrate. Therefore, it is likely that other metal species are also released during filtration. Characterization and analysis of elements with demonstrated bactericidal capacity, and which are commonly found in clays – e.g., Mg, Cu, Si, Al, Ti, Fe, Zr – is specifically recommended.

This research also clearly demonstrated a novel and valuable approach to modelling CWF performance with generalized characteristics of porous media, yet various features of CWF design were excluded from the analysis. Future work is thus required to narrow the range of optimum filter characteristics. For instance, investigation into different, particularly smaller, sieve sizes is needed. Additionally, the particle size itself was not well controlled but was added only as <595 μm and <850 μm. Investigation of narrower ranges (e.g., $500 < x < 595$),

as well as how such ranges translate to the specified media characteristics, is recommended. Finally, evaluation of criteria such as mechanical clay properties, firing process, or firing material type would be beneficial. Inclusion of more data from diverse settings may also reduce the uncertainty observed therein.

With respect to implementation, this research found that health outcomes were positively correlated with certain drinking water sources during different time periods, suggesting CWF performance was not entirely consistent. Future work should quantify how CWF disinfection efficacy in the field relates with variable influent water qualities. Further, as demonstrated by the technical research from Chapter 4, this quantification should also include non-microbial water quality parameters such as dissolved oxygen, pH, and turbidity, which are often excluded from such analyses in literature. With that said, I believe an alternative to household-scale CWF deployment and behaviour change programming requires exploration.

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Appendices

Appendix A - Supplementary Material (Chapter 4)

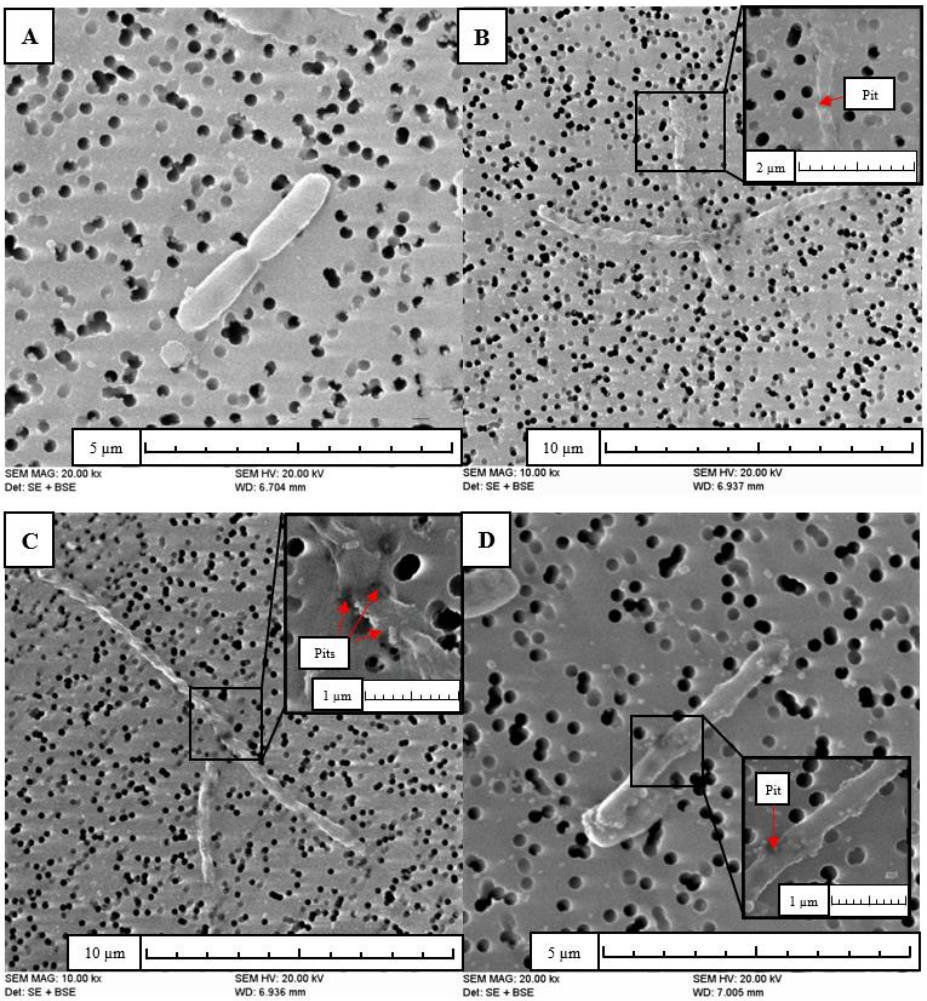

Figure A.1 Scanning Electron Microscopy Images of (A) an undisrupted E. coli cell, (B) a lysed E. coli cell after treatment with 1 mg/L Ag (Inset: Pit formed from AgNP disinfection), (B) a lysed E. coli cell after treatment with Ag67ZnO33 (Inset: multiple pits formed by AgNP/ZnO mixture), and (D) a damaged E. coli cell after treatment with ZnO1 (Inset: pit formation after ZnO1 disinfection)

Appendix B - Preliminary Results from Two Additional Groups of Women in Longido

B.1 Introduction

Presented in this appendix is the preliminary analysis and discussion of data related to CWF usage, water usage, knowledge retention, and associated health impacts among two additional groups of participants within the Longido District in Tanzania. Specifically, this section discusses two filter intervention studies among women recruited in partnership with the Longido District Health Clinic (LDHC) and women already engaged in TEMBO's adult literacy program in the Oldorko village of Orbomba Ward. These interventions consisted of filter provision and knowledge communication and engagement programming that differed from the methods applied in Kimokouwa (Chapter 7) only in terms of duration on repetition (see Appendix B.2.2). Please note some results shown in Chapter 7 are also presented again herein for comparative purposes. The primary objective of this section is to evaluate the long-term relationship between knowledge communication and participant engagement strategies with WaSH behaviours, WaSH information retention, and the diarrheal health of women and their children in Longido, Tanzania, after receiving a CWF.

B.2 Methods

The community consultations and associated collaborative program development strategy described in Chapter 7 were similarly employed herein. Participants in this study were thus presented identical WaSH information and asked the same survey questions (Appendix C).

B.2.1 Recruitment

Sixty-five women were recruited to this study through either TEMBO's Adult Literacy Program in Oldorko (41 participants) or the LDHC (24 participants). In the latter group, women attending child and maternal health services between June and July 2019 were invited to participate by a residing healthcare professional if they (1) had a child under one year of age or was going to give birth within the upcoming year, (2) lived within a 30-minute driving radius of the LDHC, and (3) did not already have access to a non-consumable (e.g., chemical disinfectants, coagulants) drinking water treatment technology. The intention behind these selection criteria was to facilitate improved access to safe drinking water particularly for children within the first 1000 days of life, as this is the most vulnerable period in which diarrheal illness may lead to mortality (GBD Risk Factor Collaborators 2020, WHO 2017). Note that individual selection was conducted exclusively by a nurse or doctor and no individual was contacted by the research team until after consultation with a healthcare professional. Moreover, patients were subsequently directed to attend an initial consultation meeting with the research team, during which details on the study purpose and procedure were provided. If the individual was interested in participating, they were asked to read and sign a consent document (available in English or Swahili) to demonstrate their understanding of the program and the information being requested of them. If the individual preferred, this consent document was also presented orally and/or in Maa (the local Maasai language), and they could provide a thumbprint instead of a signature. After prior and informed consent was provided, participants were presented with the filter package and associated education program, as described in Appendix B.2.2.

Members of the Oldorko Literacy Group were already engaged in the literacy program at project inception. Initial engagement began in March 2021 when the research team member and local water champion, "T", attended a group meeting after invitation by TEMBO staff and literacy program leaders. During this initial meeting, T provided group members with a detailed account of the study information, including its purpose, procedure, and the information that would be requested of them. T then read the related consent document to each group member individually in Maa, after which they were requested to sign the document or provide a thumbprint as demonstration of understanding. Consent was obtained from every member of the literacy group before knowledge communication began. These procedures were approved by Carleton University Research Ethics Board (CUREB) and the Tanzanian Commission on Science and Technology (COSTECH).

B.2.2 Implementation

Shown in Figure B.1 are the intervention schedules for each study group evaluated. As observed, engagement with Health Clinic and Kimokouwa Literacy Groups began in July 2019 and extended until April 2022. The implementation program among the latter group is described in detail in Chapter 7. After the consent procedure was completed among the former group, participants were invited to undergo a baseline interview and attend an education session soon thereafter. More specifically, each participant was first requested to provide information related to their socioeconomic status and water usage practices. Knowledge related to vectors for disease transmission, filter care and maintenance, and other relevant topics was then communicated to between one and five participants at the LDHC on the same day as their recruitment (See Appendix C). After this session, contact information

was requested and a filter package delivery was scheduled within the upcoming week, as per the participants' convenience. The same knowledge communication program was then repeated for a second time with the participant at their home before the filter package (filter, bucket, stand, cleaning supplies, educational pamphlet (Appendix C), notebook, and two pens) was provided. Each of these two knowledge communication sessions were approx. 60-75 minutes in duration. Follow-up interviews were subsequently conducted at participant homes 3, 6, 12, 15, 18, 21, 24, and 27 months after filter and program provision.

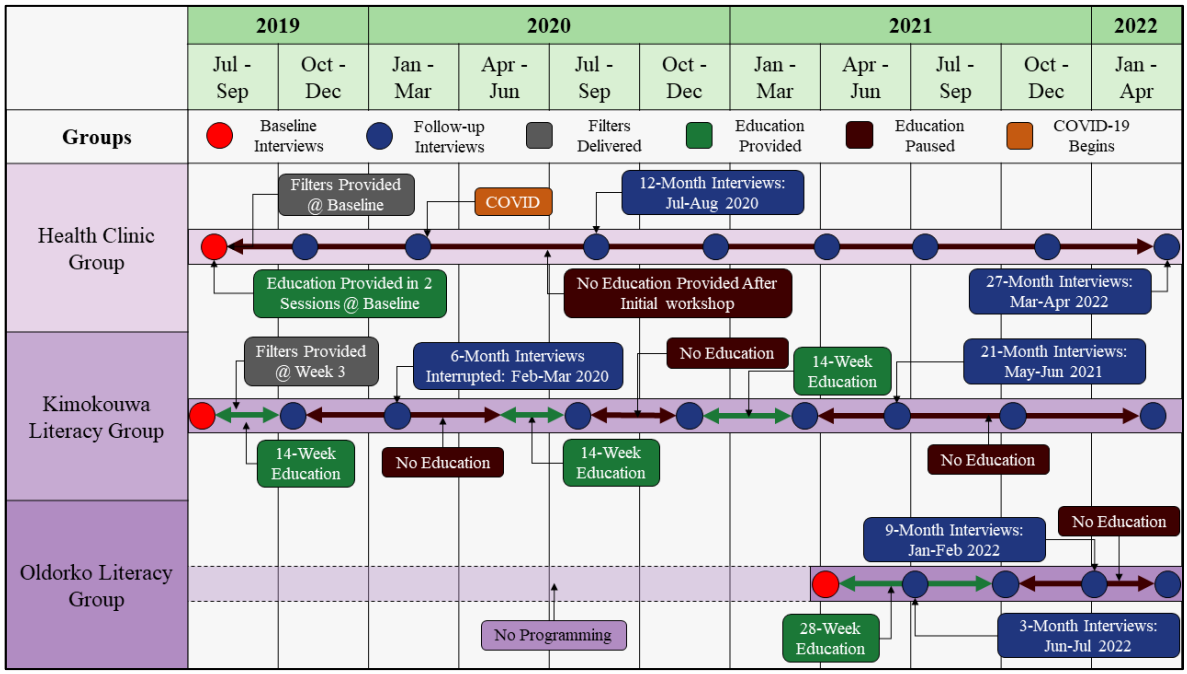

Figure B.1 Intervention timelines across all three groups

After consent was affirmed among members of the Oldorko Literacy Group in March 2021, baseline interviews were conducted over the following two weeks. Upon completion, knowledge communication programming began in April 2021 and extended for 28 weeks thereafter. The program was divided into 7 modules, which were each twice repeated

consecutively for a total of 14 weeks (the same program as was provided in Kimokouwa). The entire 14-week program was then repeated again, extending to the end of September 2021. Two review lessons were also conducted after the second round of programming. All knowledge communication was subsequently paused, and follow-up interviews were conducted at the participants' homes after 6, 9, and 12 months from the time of filter provision. Filters were delivered to participant homes after the third week of programming. Each lesson in Oldorko was approximately 10-15 minutes in duration. Data collection in Oldorko is ongoing and scheduled to continue until 2023.

All knowledge communication was completed using a participatory education structure. Namely, dialogue was highly encouraged throughout each session between the project's local water champion and study participants. As such, while specific information and learning objectives structured the engagements, examples and implications were identified with significant participant involvement such that the link between the information and their lived experiences was evident. All elements of program implementation were further completed by the project's local water champions in partnership with TEMBO's community engagement team, who are all also local Maasai women.

B.2.3 Data Management and Analysis

All data was collected by the project water champions using the mWater smartphone app under Wine to Water (W2W) International's licence and stored on mWater's secure servers. Child health was further reported by their mothers; no children were contacted directly. For analysis, data was downloaded from their online dashboard as a .csv file and stored on a

password-protected hard drive. Data was then cleaned and organized in MS Excel and visualized and analysed in RStudio. Chi-squared tests were conducted to determine the differences between groups and logistic regression analysis was used to determine the relevance of various measured factors on reported diarrheal outcomes among participating women and their children.

B.3 Preliminary Results and Discussion

B.3.1 Reported Diarrheal Outcomes

The reported diarrheal outcomes (darker blues) among participants and their children are illustrated in Figure B.2. A first important observation is that diarrhea was relatively widespread across all three groups at baseline. That is, 37% of adults and 33% of children reported regularly experiencing diarrhea, where 'regular' was defined as more than once per month. No significant difference was observed between adults and children ($p=0.5$), demonstrating the relevance of water-related illness to all age groups in these communities. With that said, the differences in overall prevalence (i.e., adults and children combined) between groups were statistically significant ($p<0.05$). More specifically, 26% of all adults and children (combined) in the Kimokouwa Literacy Group (KLG) were reported to regularly experience diarrhea, while 42% reported the same within both the Health Clinic Group (HCG) and Oldorko Literacy Group (OLG). Less diarrheal illness was thus observed within Kimkokouwa than their neighbouring communities, highlighting how water-related illness is dynamic across time and space within a given geography and between populations.

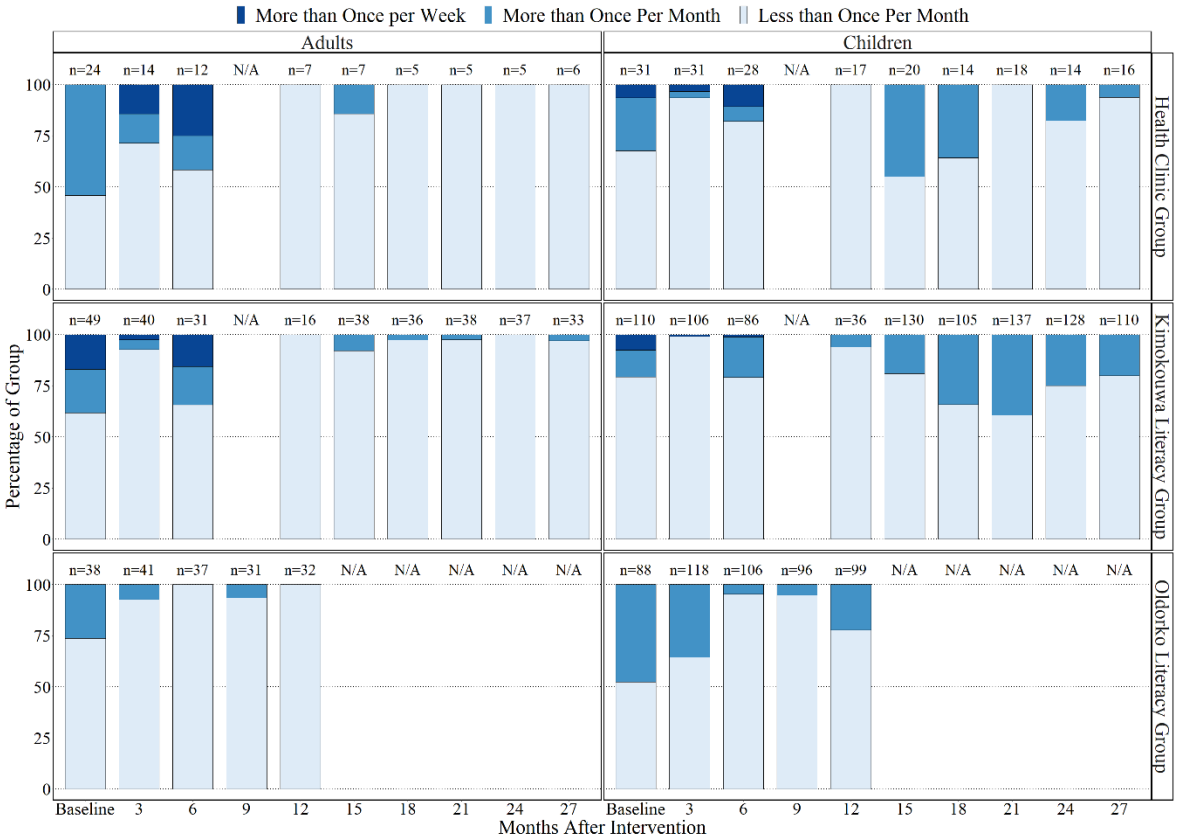

Figure B.2 Diarrheal illness among participants and their children over time per group

3 months after filter distribution, 4, 3, and 3 participants in HCG, KLG, and OLG reported experiencing diarrhea, corresponding to 29%, 8%, and 7% of the groups, respectively. The percentage of each group reporting diarrhea at the 3-month follow-up was thus consistently less than at baseline, though a larger proportion of interviewed members of HCG reported illness than those from KLG or OLG. The significance of this difference was, however, impossible to confirm due to the reduced statistical power caused by the large number of participants from HCG who dropped out of the project within the first 14 weeks (See B.3.2.1). Regardless, some impact related to the distribution of CWFs is observed, particularly within the literacy groups.

The differences between groups and intervention strategies became more evident thereafter. That is, at the 6-month follow-up, 35% and 42% of participants within KLG and HCG reported illness, respectively, while no illness was reported among any adults in OLG. Similarly, 20% and 21% of children were reported to have experienced regular illness within KLG and HCG, respectively, while less than 5% were reported to have experienced the same within OLG. These results thus appear to confirm the hypothesis detailed in Chapter 7 that diarrheal illness was correlated with knowledge communication and program engagement. Namely, OLG had consistently received educational programming over the 30 weeks prior to these interviews, presumably resulting in high rates of filter usage and maintenance, as well as practice of other WaSH-related activities discussed that led to improved health (see Appendix B.3.2). Meanwhile, after programming was paused for KLG (between 3- and 6-month interviews), reported illness increased to near baseline levels for both adults and children. Within HCG, though fewer individuals reported illness at the 6-month interviews than at baseline, the difference between these follow-up periods was insignificant ($p=0.9$) and the reported severity of illness increased as well. The impact of the intervention and associated knowledge communication may have thus been reduced by virtue of the long duration between filter and knowledge provision and this follow-up.

Selection bias is also considered relevant within HCG results throughout the entire intervention period, and particularly after these 6-month interviews. Namely, as highlighted by Table B.1, a much larger percentage of filters distributed to HCG broke than within the other groups. Those whose filters remained were thus likely those who used, cleaned, and protected them with the greatest enthusiasm, potentially producing biased results towards

improved health than may be expected from the broader group. This hypothesis is supported by the observation that none of the 5-7 participants from HCG with working filters between 12- and 27-month interviews reported illness at any point throughout the study period (other than at baseline). The improved health reported during these latter interviews may furthermore be responsive to the intervention's success among these individuals rather than the group in general.

Conversely, reported diarrhea also remained low among participants in KLG and OLG for the duration of the follow-up schedules, while the number of participants interviewed remained representative of the broader groups. The improvements in health therein are thus hypothesized to have resulted from the knowledge communication programming in concert with the associated benefits of CWF usage. Namely, the educational programming is believed to have facilitated an improved understanding of filter care procedures and WaSH-related behaviours that translated to a higher proportion of participants maintaining such practices over time, improving health by extension. This hypothesis is supported by observations detailed in Section B.3.2.

With that said, the same long-term impacts were not observed among children in any group. For instance, between 12- and 27-month interventions, 25% of children reported illness compared with 3% of adults. Children were thus 3.2 (CI: 2.2-4.7; $p<0.01$) times more likely to report illness after intervention than adults when all other measured social and environmental factors were considered. As also described in Chapter 7, the present intervention was thus evidently challenged in equally meeting the water-related needs of

children. Broader programming is required to address the various vectors of WaSH-related illness across a community landscape and advance positive outcomes among all individuals.

B.3.2 Social & Environmental Factors of Health

B.3.2.1 Filter Usage and Breakage

Perhaps the most powerful outcome observed within this research is how filter breakage and usage differed between groups over the study period. Namely, as detailed in Table B.1, 50% (12) of all filters distributed to HCG had broken by the 6-month mark, whereas 18% (9) and 0% had broken among members of KLG and OLG, respectively. Further, by the 12-month mark, the number of broken filters among HCG participants increased to 67% (16), compared with 22% (11) and 12% (5) within KLG and OLG, respectively. By the final follow-ups with HCG and KLG members, 75% (18) and 24% (12) were broken, respectively. Members of HCG were thus evidently more challenged in ensuring filters were sufficiently protected to avoid breakage than their counterparts in KLG and OLG. Further, with no broken filters observed among OLG until the 9-month follow-up, the 28-week knowledge communication program appears to have improved filter care among these participants when compared with those in the other groups.

Table B.1 Filer usage and breakage over time among members of all three study groups

Topic	Months After Intervention	Health Clinic Group	Kimokouwa Literacy Group	Oldorko Literacy Group	Total
Filters in Households (%[1])	3	14 (58%)	43 (86%)	41 (100%)	98 (85%)
	6	12 (50%)	41 (82%)	41 (100%)	94 (82%)
	9	--	--	37 (90%)	--
	12	8 (33%)	39 (78%)	36 (88%)	83 (72%)
	15	7 (29%)	38 (76%)	--	45 (61%)
	18	6 (25%)	38 (76%)	--	44 (59%)
	21	6 (25%)	38 (76%)	--	44 (59%)
	24	6 (25%)	38 (76%)	--	44 (59%)
	27	6 (25%)	38 (76%)	--	44 (59%)
Filter in Use[2] (%[3])	3	11 (78%)	28 (70%)	41 (100%)	80 (84%)
	6	9 (75%)	30 (94%)	36 (97%)	75 (93%)
	9	--	--	29 (93%)	--
	12	5 (71%)	16 (100%)	32 (100%)	53 (96%)
	15	6 (86%)	38 (97%)	--	44 (98%)
	18	4 (80%)	36 (100%)	--	40 (98%)
	21	5 (100%)	38 (100%)	--	43 (100%)
	24	4 (80%)	36 (97%)	--	40 (95%)
	27	5 (83%)	33 (100%)	--	38 (97%)

[1] % of filters in households represents percentage of filters remaining in households relative to the number distributed
[2] Filter in Use refers to the number of filters with water either in the bucket or in the pot at the time of interview
[3] % of Filters in Use represents the percentage of filters in use relative to the number of individuals interviewed at that follow-up period in each group

The relevance of the intervention strategy is further notable within the number of filters that were observed as in use at the follow-ups, as well as the reason why filters had broken. Specifically, 80% (49/61) of participants in HCG had water present either within the filter or receptacle at the follow-up interview time, compared with 94% (255/272) and 98% (138/141) of participants in KLG and OLG, respectively. And as shown in Figure B.3, 78% (14/18) of filters that broke among members of the HCG were caused by a participants' child, while 46% (6/13) and 40% (2/5) broken for the same reason within KLG and OLG, respectively. These results together therefore suggest that participants in KLG and OLG may have been

more inclined to use their filters, as well as more protective over them, resulting in lower proportional breakage and associated improvements in diarrheal health (particularly among the adults).

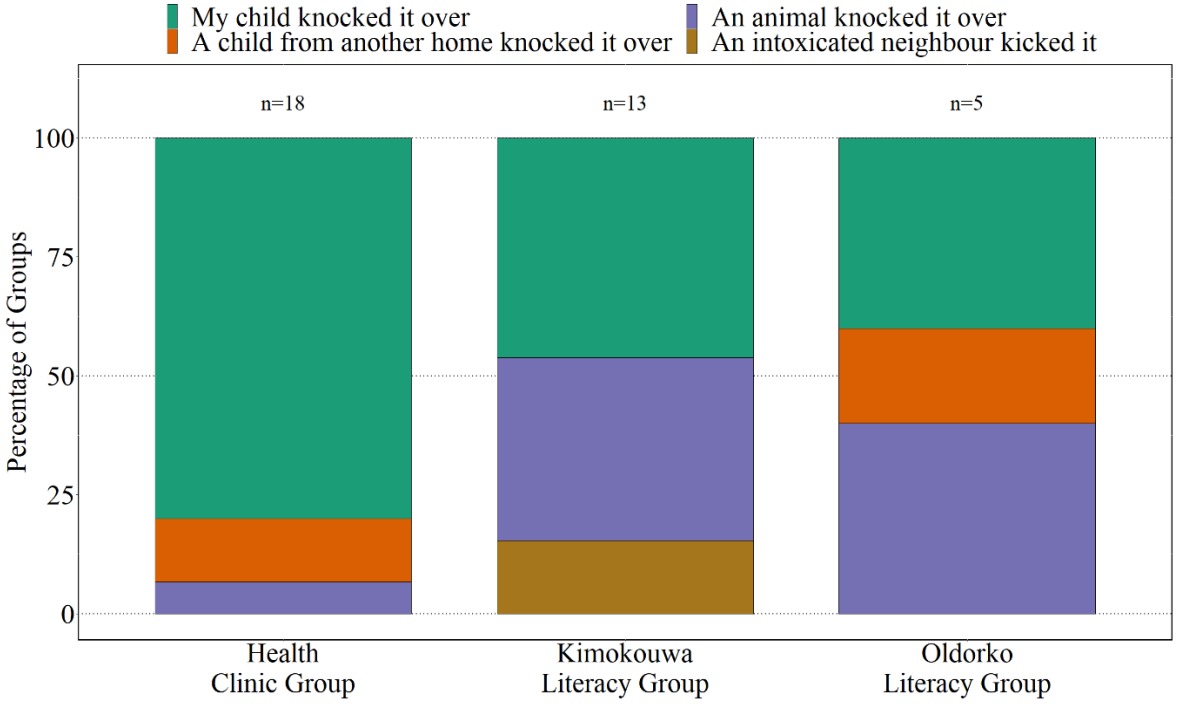

Figure B.3 Reasons for broken filters in each group

B.3.2.2 Water Sources

Previous research has highlighted that filter efficacy may change at different times in the year due to changing water sources and water quality changes within those sources (Dias, et al. 2018, Casanova, et al. 2012, Francis, et al. 2015). These observations are also supported by the technical explorations in Chapter 4 that found water quality, and particularly dissolved oxygen, impacts CWF efficacy. The diarrheal health results presented in Appendix B.3.1 may thus be further clarified with investigation of water source. Table B.2 details the percentage of participants from each group that used each water source over time. The

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binomial regression analysis further demonstrated that no water source significantly contributed to reported illness more than any others at baseline, though using a surface water source resulted in a 1.5 (CI: 1.1-2.2; p=0.02) times higher probability of reporting illness after intervention. No significant impact could be predicted based on consumption from any other sources. The times when surface water was consumed in the largest proportions was further at 3- and 21-month follow-ups, when diarrhea was particularly low among adults but higher among children (especially in OLG). The relevance of surface water consumption on health is thus predominantly driven by the results among children. It is, however, impossible to determine the cause behind this correlation without water quality estimates from before and after filtration with this source. For instance, surface water sources are colloquially known to be more contaminated and turbid than others in the community, suggesting the spike in illness may be related to drinking it directly rather than after filtration. This consideration is also supported by previous studies demonstrating that filters remove more bacteria when suspended solids concentrations are higher (Abebe, Chen and Sobsey 2013). Alternatively, due to the higher contaminant loads, effluent concentrations may still have been high risk and thus yet too high to prevent illness among those most vulnerable. More data is required on CWF field performance with differing water sources, as well as child behaviour related to CWF usage in the home.

Table B.2 Drinking water source usage over time among each study group

Water Source	Months After Intervention	Health Clinic Group	Kimokouwa Literacy Group	Oldorko Literacy Group	Total
Dam	0	8 (33%[1])	19 (39%)	10 (26%)	37 (33%)
	3	2 (14%)	5 (12%)	0 (0%)	7 (7%)
	6	3 (25%)	5 (16%)	6 (16%)	14 (17%)
	9	--	--	4 (13%)	--
	12	0 (0%)	5 (31%)	0 (0%)	5 (9%)
	15	1 (14%)	33 (87%)	--	34 (76%)
	18	0 (0%)	1 (3%)	--	1 (2%)
	21	0 (0%)	1 (3%)	--	1 (2%)
	24	0 (0%)	4 (11%)	--	4 (10%)
	27	0 (0%)	6 (18%)	--	6 (15%)
Well	0	6 (25%)	26 (53%)	0 (0%)	32 (29%)
	3	1 (7%)	22 (53%)	0 (0%)	23 (24%)
	6	0 (0%)	0 (0%)	0 (0%)	0 (0%)
	9	--	--	0 (0%)	--
	12	1 (14%)	1 (6%)	5 (16%)	7 (13%)
	15	0 (0%)	2 (5%)	--	2 (5%)
	18	0 (0%)	0 (0%)	--	0 (0%)
	21	0 (0%)	4 (10%)	--	4 (9%)
	24	1 (20%)	0 (0%)	--	1 (2%)
	27	1 (17%)	2 (6%)	--	3 (8%)
Rainwater	0	4 (17%)	6 (12%)	0 (0%)	10 (9%)
	3	0 (0%)	8 (19%)	0 (0%)	8 (8%)
	6	5 (42%)	12 (38%)	0 (0%)	17 (21%)
	9	--	--	3 (10%)	--
	12	0 (0%)	1 (6%)	3 (9%)	4 (7%)
	15	0 (0%)	0 (0%)	--	0 (0%)
	18	0 (0%)	0 (0%)	--	0 (0%)
	21	0 (0%)	0 (0%)	--	0 (0%)
	24	4 (80%)	15 (40%)	--	19 (45%)
	27	1 (17%)	0 (0%)	--	1 (3%)
Surface Water[2]	0	3 (13%)	30 (61%)	0 (0%)	33 (30%)
	3	5 (36%)	13 (31%)	23 (56%)	41 (42%)
	6	2 (17%)	14 (44%)	0 (0%)	16 (20%)
	9	--	--	2 (6%)	--
	12	0 (0%)	0 (0%)	2 (6%)	2 (4%)
	15	0 (0%)	5 (13%)	--	5 (11%)
	18	4 (80%)	1 (3%)	--	5 (12%)
	21	1 (20%)	38 (100%)	--	39 (91%)
	24	3 (60%)	6 (16%)	--	9 (21%)
	27	0 (0%)	0 (0%)	--	0 (0%)

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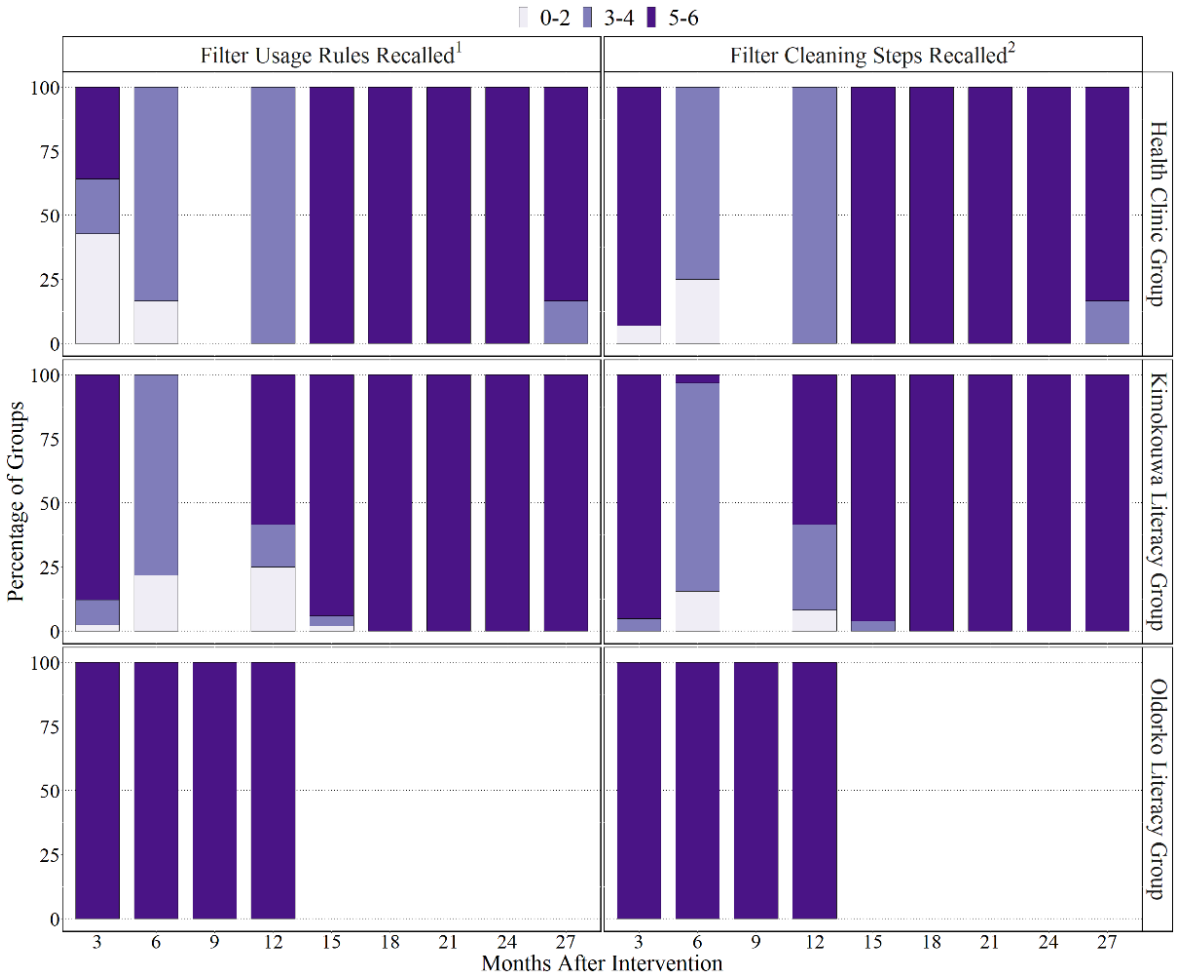
Village Tap	0	18 (75%)	32 (65%)	37 (97%)	87 (78%)
	3	8 (57%)	20 (48%)	41 (100%)	69 (71%)
	6	10 (83%)	21 (66%)	37 (100%)	68 (84%)
	9	--	--	30 (97%)	--
	12	5 (71%)	7 (44%)	32 (100%)	44 (80%)
	15	6 (86%)	38 (100%)	--	44 (98%)
	18	5 (100%)	36 (100%)	--	41 (100%)
	21	5 (100%)	2 (5%)	--	7 (16%)
	24	4 (80%)	25 (65%)	--	29 (69%)
	27	5 (83%)	27 (82%)	--	32 (82%)

[1] % refers to the percentage of interviewees reporting using each water source at the follow-up time per group. Participants often use multiple water sources at a time so percentages may not add to 100

[2] Surface water refers to water collected from either streams that naturally form from mountain runoff (wet season), or human-made rainwater retaining reservoirs (dry season)

B.3.2.3 Knowledge Retention

A final intervention measurement in need of evaluation is the retention of key learning objectives across groups. Specifically, the number of filter protection and cleaning rules recalled by participants is presented in Figure B.4. A first important observation is that knowledge retention by participants in OLG was consistently near-perfect throughout the entire study period, highlighting the value of the knowledge communication program therein. Meanwhile, participants in HCG recalled a significantly lower number of rules and/or cleaning steps within the first twelve months than members of both literacy groups ($p<0.05$). The complete recall within this group in the months thereafter is further considered a result of the selection bias detailed above (Section B.3.1). With that said, while recall of filter rules was not significantly related to reported illness ($p=0.09$), participants were 1.5 (CI: 1.15-1.85; $p<0.01$) times less likely to report illness for every cleaning step recalled. Proper filter cleaning and maintenance is thus demonstrably important to associated diarrheal health. This result further supports similar observations found by Meierhofer et al. (2018).



[1] Filter usage rules include (1) use a clean cup, (2) no playing near the filter, (3) be gentle with the tap, (4) do not touch the tap opening, (5) do not touch the outside of the filter, (6) leave the filter in the bucket (unless cleaning)

[2] Filter cleaning steps include (1) clean the bucket and lid with soap and water, (2) scrub the inside of the filter with a brush and boiled water, (3) remove grimy residue with boiled water and dump, (4) scrub the outside of the filter with brush and boiled water, (5) pour boiled water over entire filter, (6) enclose filter within cleaned bucket and lid

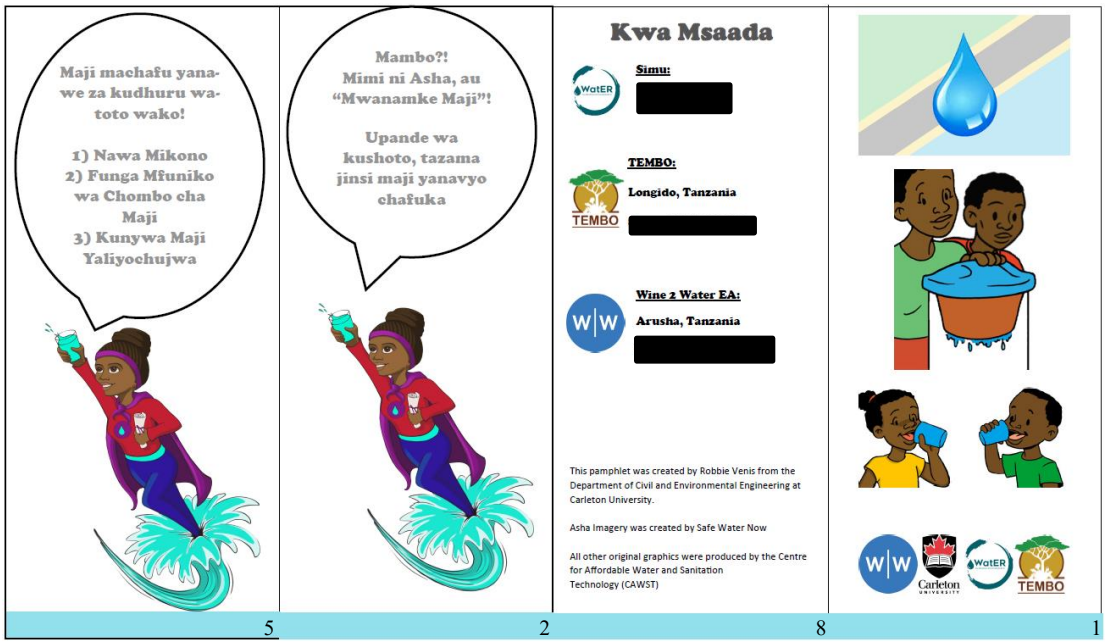
Figure B.4 Number of filter usage rules and filter cleaning steps recalled by members of each group over time

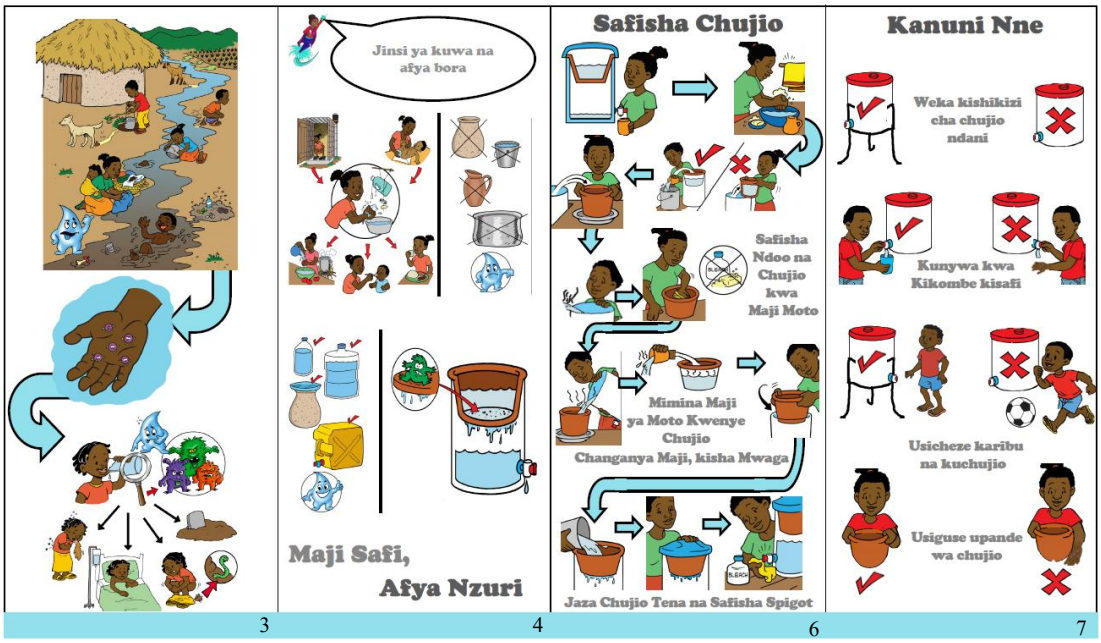
B.4 Conclusions

Knowledge communication and participant engagement programming appears significantly related with diarrheal health outcomes among women in Longido, though impacts were weaker among children. The greatest effects were further observed in terms of reducing filter breakage. More research into drinking water behaviour and water quality is needed.

Appendix C - Chapter 7 Supplementary Material

Pictorial Education Pamphlet





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C.1 Education Program

<u>WASH Activity 1: How Water Gets Dirty</u>

Before giving out the pamphlet, ask women to provide examples of how water can become dirty

Ask women if they know of something they did today that could have made water dirty

Examples:

1) pooed or peed in the bush

2) washed clothes near water

3) changed a baby's diaper near water

4) threw garbage into water source or on ground near water source

Explain today you will talk about how this can make water dirty

Hand out pamphlet and talk about first picture ONLY:

Ask women to identify different ways water becomes dirty in the first picture, and explain why that makes it become dirty:

1) Runoff from farming (#1)

Chemicals are used on farms and in gardens to make plants grow. When it rains, those chemicals are washed into water, which is not good to use or drink.

2) Animal poo and pee (#2)

Inside animal poo and pee are tiny, invisible creatures that make people sick if they eat of drink them, called microorganisms. When animals poo and pee on the ground, those creatures can get into water, which makes us sick when we consume it

3) Human poo and pee (#3)

This is the same as animal poo and pee. Human poo and pee can also hurt our animals.

4) Garbage (#4)

When garbage is on the ground, rain, people, or animals can move it to the water source, which can make the water dirty. When we then drink water with garbage in it, it can make us and our animals sick.

5) Cleaning clothes and dishes (#5)

Clothes and dishes have lots of dirty things on them from being used, which can get into water when washing. Even if washing is done away from the water, rain will still move those dirty things into water. When we drink that same water, it can make us sick.

6) swimming in water (#6)

Just like washing, all of the dirt on your body will get into the water if you go into it, which can make us sick if drinking or washing with it

Ask women to think of other ways that water becomes dirty

Examples:

1) people/animals drinking directly from a tap

2) using water while having dirty hands

3) grease and oil from cars

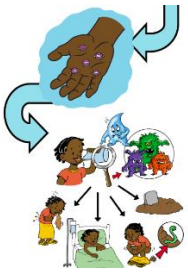
4) …?

General Point to remember for activities near water:

When people even go near water, it becomes possible for that water to become dirty. An example would be that somebody stepped in pee or poo while walking and brought those creatures (microorganisms) to the water with them, making it dirty. Or, if somebody has dirty hands and puts them in the water, that water will become dirty too. As more people and animals use the same water source, it becomes more likely that the water is dirty, and that using it will make us, our children, and our animals sick.

<u>WASH Activity 2: How Dirty Water Can Make Us Sick</u>

Hand out pamphlet to everybody and point to the 2nd and 3rd pictures on the first panel.



Explain that the small dots on the hand are called MICROORGANISMS. These are the tiny creatures that cause problems like typhoid or cholera. They get into the water or on hands by having contact with human or animal poo. This can be by directly touching the poo, or by touching a surface that had poo on it, like an animal, water jug, or anything else.

Explain that bacteria live in air as well as water. This is why you must cover your mouth when you cough or sneeze. Because when you sneeze or cough, you can spread the disease onto your hands or onto the person next to you.

Point to the pictures on the second panel of the pamphlet with the water containers covered and uncovered. Explain that the bacteria in the air can get into the water if they are open, which is why ***it is important to cover the containers.*** *This is a helpful way to protect ourselves from diseases.*

Ask women to give examples of how those microorganisms can get onto their hands. Try to connect this information with how water gets dirty in last two weeks:

Examples:

1) after using the toilet

Microorganisms like cholera that are in our poo can touch our hands directly, or float into the air and get on our hands. This is why we MUST wash our hands after using the toilet.

2) after milking animals

Animals have many microorganisms all over their bodies. Every time we touch them, we risk those microorganisms getting onto our hands and then into our bodies when we eat or drink.

3) after changing a child after pooing

Just like in our poo, microorganisms are in our baby's poo too. When we change a baby, we often touch their poo, and those microorganisms get onto our hands.

4) after sneezing or coughing

Just like in our poo and pee, microorganisms live in our mouths and noses. When we sneeze or cough, those microorganisms go into the air and make everything around us dirty. This means that if we sneeze or cough into our hands, they will become dirty

Next, explain when hands are dirty, everything they touch becomes dirty too. This is how disease spreads from person to person.

This means when hands touch water, that water becomes dirty and can make you sick

When we drink dirty water, our bodies have to fight against the diseases. If there are more diseases than strength in our bodies, we get sick.

This is why children get sick more than adults – they do not have as much strength as adults. This is why drinking clean water is most important for children, even though it is important for everybody

Using the filters:

This is also why it is good to use the filters. The filters will remove the microorganisms from our water, making it clean to drink and safe for us and our children.

Point to 4th picture, where the filter can be seen on the second panel:

To use a filter, just pour the water into the top and wait for it to drip out of the bottom. ***You must also let water go through the filter one time before you can drink from it.***

The water will also only be safe if you follow these rules:

1) no touching water in the bucket

2) no touching the filter pot unless cleaning

3) always use a clean cup to drink from the tap. Never drink from filter with your hand!

4) never touch the tap on the bucket.

We will talk more about the filters in the next few weeks, including how to keep it safe, how to make sure you are always drinking clean water, and how to clean it.

WASH Activity 3: Why we wash hands, how we wash hands,

<u>NOTE: BRING A BUCKET OF WATER WITH SOAP TO CLASS FOR DEMONSTRATION</u>

Hand out pamphlet and ask women to look at first picture on second panel

Ask women to point to all of the times when we should wash our hands:

1) after using the toilet
2) after changing a baby
3) before cooking
4) before feeding a child
5) before eating

Ask women to say any other times when they should wash their hands

1) after milking an animal
2) after doing cleanness
3) …other examples from the women…

Ask women to explain why we wash our hands.

<u>Reasons that should be said:</u>

1) to remove microorganisms from our hands
2) to prevent diseases from entering our bodies when our hands our dirty
3) to prevent diseases from entering other people's bodies when our hands are dirty

Point to woman washing hands in pamphlet and demonstrate how to wash hands (use bucket and soap that you brought with):

1) pour water onto hands to make them wet
2) rub soap onto hands
3) make sure to rub soap on top of hands, palms of hands, and between the fingers
4) rub soap all over hands for 20 SECONDS
5) rinse soap off of hands with water
6) dry hands on a clean cloth or in the air

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Ask women to volunteer to wash their hands and have them do the same thing as you. At least 3 women should volunteer to wash their hands in the class.

Once handwashing is done, review how to use the filter. Explain that you put water in the top and wait for it to drip out of the bottom.

Make sure to say rules to keep water safe, including:

1) no touching water in the bucket

2) no touching the filter pot unless cleaning

3) always use a clean cup to drink from the tap. Never drink from filter with your hand!

4) never touch the tap on the bucket.

Keeping the filter safe:

Before finishing, point to 4th panel of pamphlet to show the rules of keeping the filter safe. Explain that filters are very fragile and will break very easily. This means that it is very important to keep the filter in a safe place and to treat it with respect. The most important rules to follow to keep the filters safe is:

1) ALWAYS keep the filter in the stand unless you are cleaning it

2) Be VERY gentle with the tap.

3) Do not let children play near the filter

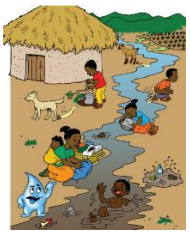
4) Do not let animals near the filter

WASH Activity-4: WEATHER and WASH IMPORTANCE

Ask women if there is any time of year that they notice more people get sick. Ask them if people get sick during the rain and flood season

Next ask, based on what was learned in previous weeks, if women can explain why rain season makes more people sick

After asking these questions, hand out pamphlets again and point to first picture on first panel. Explain again all of the ways that water gets dirty (from first lesson).



Explain next that during rains, all of the things in that picture are carried into the water by the floods. When this happens, the water gets dirtier than during other times in the year, which is why more people get sick.

Also explain that when floods happen, the tiny diseases are spread all over the land, meaning diseases are in more places than during dry times.

This is also why we use the filters, which we all now have. Because they remove those diseases from the water, making the water clean.

Review again how to use the filter. Explain that you put water in the top and wait for it to drip out of the bottom, like in this picture:

Make sure to say rules to keep water safe, including:

1) no touching water in the bucket

2) no touching the filter pot unless cleaning

3) always use a clean cup to drink from the tap. Never drink from filter with your hand!

4) never touch the tap on the bucket.

<u>WASH Activity 5: TOILETS</u>

Ask women how many of them defecate in the bush.

Ask women how many of them have access to a toilet.

Ask them if they can tell you the different types of toilets:

1) Flush toilets with water

2) latrine toilets

Ask if any of them can explain to you why people would use a toilet instead of just going in the bush. Tell them to think about what we have talked about in the past weeks.

Explain:

1) It is good to use toilets because they keep the poo and pee off of the ground

2) When the poo and pee is off the ground, rain cannot move it

3) this means it is less likely that poo or pee will enter the water source or your homes

4) it is always safer to poo or pee in a toilet than in the bush

Explain that if the women do not have access to a toilet, building one can help protect us, our families, and our animals from diseases, especially when it rains. If you cannot afford to build a toilet, it is good to

1) speak to neighbours in your boma and nearby bomas to work together to pay for a toilet together. Sharing a toilet is better than having none.

2) Speak to your community leaders and explain that you need a toilet. Ask village leaders to advocate for your rights.

If you cannot get access to a toilet at all and have to go in the bush, there are some ways that can still help protect us against diseases, though they are not as good as toilets. Ask the women if they can think of any:

1) Pee and poo far away from any homes

2) Pee and poo far away from any water sources

3) Pee and poo far away from the market, or any other place where many people gather

4) Be very careful not to get pee or poo on your hands, feet, or shoes

5) When you pee or poo in the bush, it is better to dig a small hole, pee and poo into the hole, and then cover the hole with dirt and pack the dirt down on top. This keeps poo away from the surface of the ground and can help prevent diseases from being picked up by rain and getting into our water.

Explain that the filter cleans the water and makes it safe, but this is our last defense against disease. It is always better to stop the amount of disease that gets into our water before we have to treat it with the filter.

WASH Activity 6: How the filter cleans water

Ask women why they think the filters are good. Try to get women to explain:

1) filters remove microorganisms from water

2) Microorganisms make us sick when they get into our bodies

Ask if any of them know how the filters make water clean. Ask them to give best guess if they don't have an answer

Hand out the pamphlets to the class

Point to the picture of the filter on the second panel of the pamphlet. Show them how the water is dirty before filtering, but clean afterwards.

Explain that the filter has tiny holes that are so small that diseases like typhoid and cholera (microorganisms) cannot get through them. This is why the inside of the filter gets more and more dirty the more that you use it, and why you have to clean them.

Explain how using cloth to filter water is similar to ceramic filter, but the holes in a cloth are bigger than the ceramic filter, so less microorganisms are removed. This is why it is better to keep using the ceramic filter.

NOTE: if there Is there an example that women are familiar with that could be used to explain this idea more, please explain it to the group

Review, again, how to use the filter, how to keep the water safe, and how to keep it safe from breaking.

Make sure to say rules to keep water safe, including:

1) no touching water in the bucket

2) no touching the filter pot unless cleaning

3) always use a clean cup to drink from the tap. Never drink from filter with your hand!

4) never touch the tap on the bucket.

Make sure to say the most important rules to follow to keep the filters safe from breaking:

1) Do not let children play near the filter

2) Do not let animals near the filter

3) ALWAYS keep the filter in the stand unless you are cleaning it

4) Be VERY gentle with the tap.

WASH Activity 7: How to clean the Filter

NOTE:

BRING TO THIS CLASS:

1) A FILTER IN A BUCKET

2) BOILED WATER AND COOL WATER (ASK TEMBO TO PROVIDE A FULL FLASK WITH BOILED WATER BEFORE LEAVING)

3) SOAP

4) A BRUSH

Hand out pamphlet and do a demonstration. Make sure to have women follow along with the pictures in the pamphlet. Stop and point to each picture and show what it looks like to do it in person. Ask for a volunteer to each go through cleaning process after it has been done once.

Steps to say:

1) Boil water

2) Wash hands with soap and water

3) Scrub the inside and outside of the lid with soap and water

4) wash soap off of lid with boiling water and place upside down on ground

5) Remove the filter from the bucket by the rim and place upside down on the lid

6) Scrub the inside and outside of bucket and tap with soap and water

7) Rinse the bucket with <u>boiling</u> water until all soap is removed

8) Allow boiling water to run through the tap

9) place bucket on the ground

10) rinse brush with boiling water until all soap is removed

11) turn filter over and scrub with the clean brush. DO NOT USE SOAP ON FILTER

12) pour boiling water into the filter and scrub again with the clean brush

13) swirl boiling water around in bowl to remove all dirt from inside

14) dump water out of the filter

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15) pour boiling water back into the filter again

16) swirl boiling water around in bowl to remove any remaining dirt

17) dump water out of the filter

18) turn filter upside down on top of filter lid

19) rinse brush with boiling water again until all dirt is removed

20) pour boiling water all over bottom of filter

21) brush bottom of filter from top to bottom all over

22) pour boiling water over bottom of filter again

23) using thumbs and hands, hold the filter to the lid and turn over to let water drip out of lid. MAKE SURE TO HOLD ON TIGHT AND NOT TO DROP THE FILTER!!!

24) put filter back inside the bucket carefully

25) Drink water again!

<u>WASH Review Lesson</u>

This lesson is to go over everything that we have talked about over the past few months.

We want the women to be able to explain as much of the information as possible

Only tell the women answers if they cannot answer by themselves.

1) How water gets dirty and makes us sick

Ask women to explain how water gets dirty. Ask them to give examples of things that can make water dirty. Examples:

1) pee and poo

2) garbage

3) chemicals from farming

4) other examples?

Ask why these examples make water dirty

Answer: they have microorganisms that get into the water and make us sick

Ask why rain can make water more dirty

Answer: rain picks up microorganisms from the ground and moves them to the water, making the water dirty, which makes us sick

2) How and why to wash our hands

Ask women when we should wash our hands

1) after peeing or pooing

2) after cleaning a child

3) after milking animals

4) before cooking

5) others?

Ask women why we should wash our hands

Answer: when we do these activities, microorganisms get onto our hands, and then get onto everything we touch. This means if we touch food or water with dirty hands, that food and water become dirty, and makes us sick when we eat or drink it.

Ask women what happens if we do not wash our hands

Answer: we spread diseases to each other, our families, and our animals

Ask how we wash our hands

Answer: with soap and water for 20 seconds. Wash top of hands, palms, and between the fingers

3) Toilets

Ask why toilets are better than the bush

Answer: they keep pee and poo off the ground so it cannot get into the water source or our homes

Ask how women what they can do to make going in the bush safer if they do not have a toilet

Answers:

1) go far away from the home

2) go far away from the water source

3) dig a hole in the ground and pee or poo in it instead of just on the ground

4) be very careful not to get any poo or pee on your feet or shoes

4) Filters

Ask women why it is important to use the filters

Answer: the filters remove microorganisms from water to make it clean

Ask women how to keep the filter safe

1) keep away from animals

2) keep away from children playing

3) always keep it in the stand unless cleaning

4) be very gentle with he tap

5) others?

Ask women how to keep the water from the filter clean

1) always use a clean cup

2) do not touch the tap opening

3) do not touch the water in the bottom of the bucket

4) do not touch the outside of the filter unless cleaning it

Ask women to review how to clean the filter

see steps in WaSH Activity 7

Ask women if they have any other questions about anything we learned this year. Answer any questions that they have!

C.2 Interview Questions

1. What is your name?
2. What is your marital status?
3. How many wives are you under one husband?
4. What is your religion?
5. Age:
    a. Mother:
    b. Father:
6. Activities done for money:
    a. Mother:
    b. Father
7. How many livestock do you have at your home?
    a. Cattle:
    b. Goats:
    c. Chickens:
8. How much money do you use per month? (TZS)
9. Did you attend school?
10. *If yes:* What is the last level you finished?
11. Is there a health clinic in your village?

12. Do you go to the health clinic if someone in your family is ill?
13. How often do you have diarrhea from taking medicine?
    a. More than once per week
    b. More than once per month
    c. Less than once per month
14. How often do you have diarrhea from stomach sickness?
    a. More than once per week
    b. More than once per month
    c. Less than once per month
15. Do you have children living in your home?
16. *If yes:* Details for children:
    a. Age
    b. How often do they have diarrhea from stomach sickness?
17. Where did you collect water from this month?
    a. Dam
    b. Well
    c. Rainwater
    d. Surface Water
    e. Tap Water
    f. Other (please specify)
18. How far is your home from the water source used last month?
    a. Very close
    b. Close
    c. Far
    d. Very far
19. Do you share water with your animals?
20. Is that water source clean?
    a. Yes
    b. No
    c. I don't know
    d. Somewhat (please specify)
21. Do you treat your drinking water?
22. *If yes:* How do you treat your drinking water?
    a. Filter with a cloth
    b. Chlorine/chemicals
    c. Solar radiation
    d. Boiling
    e. Decantation
    f. Charcoal
    g. Ceramic water filter
23. When do you wash your hands?
    a. After using the toilet
    b. Before eating
    c. Before feeding a child

d. After cleaning a child
e. Before preparing meals
f. After milking livestock
g. Other (please specify)

24. Why do you wash your hands?
    a. To prevent disease
    b. To remove dirt
    c. Other (please specify)
    d. I don't know
25. Do you have access to a toilet?
    a. Pit latrine
    b. Flush toilet
    c. Other (please specify)
26. *If yes:* How far is the toilet from the household?
    a. Very Close
    b. Close
    c. Far
    d. Very Far
27. *If No:* Do you defecate in the Bush?
28. How far is the defecating area from the household?
    a. Very close
    b. Close
    c. Far
    d. Very Far
29. How satisfied are you with the water filter?
    a. I don't have an opinion
    b. I don't like it
    c. I like it
    d. I like it but water collection is too low
    e. Other (please specify)
30. How often do you fill the filter?
    a. Every day
    b. Every week
    c. Every month
    d. Less than every month
31. When do you clean the filter?
    a. Every day
    b. Every week
    c. Every month
    d. Every 3 months
    e. When water filtering is too slow
    f. I have never cleaned the filter
32. Please list the rules of using the filter
    a. Always use a clean cup

b. No playing near the filter
c. Be gentle with the tap
d. Do not touch the tap opening
e. Do not touch the outside of the filter
f. Leave the filter in the bucket unless cleaning
g. Leave the filter in the stand unless cleaning

33. Please list the steps to clean the filter
    a. Clean the bucket and lid with soap and water
    b. Scrub the inside of the filter with the brush and boiled water
    c. Remove grimy residue with boiled water and dump
    d. Scrub the outside of the filter with the brush and boiled water
    e. Pour boiled water over entire filter
    f. Enclose filter with clean bucket and lid
    g. Remove all water from lid before turning right-side-up
34. *For interviewer:* is there water in the filter?
35. *For interviewer:* is there water in the bucket?

Appendix D - Other Academic Contributions

Conferences

Robbie A. Venis, Chaitanya Luhar, Onita D. Basu, 2022. The Influence of Nanoparticle-Cell Ratios on the Disinfection of *Escherichia Coli* by Silver and Zinc Oxide in Low Concentrations. Canadian Society of Civil Engineers Annual Conference. Whistler, BC [Presentation + Indexed Conference Paper]

Robbie A. Venis, Onita D. Basu, 2022. Long-Term community Engagement and Participatory Education for Improving Water and Health Outcomes: A Case Study in Rural Tanzania. Canadian Society of Civil Engineers Annual Conference. Whistler, BC [Presentation + Indexed Conference Paper]

Robbie A. Venis and Onita D. Basu, 2021. Rethinking the Implementation of Point-of-Use Water Treatment. Canadian Association of Geographers Annual Conference and Meeting. Prince George, BC [Presentation]

Robbie A. Venis and Onita D. Basu, 2021. Integrating Technical and Social Criteria for Sustained Uptake of Localized Water Treatment Technology in Rural Tanzania. International Water Associations Digital World Water Congress. Copenhagen, Denmark [Poster]

Robbie A. Venis, Virginia Taylor, Paulina Sumayani, Marie Laizer, Onita D. Basu, 2021. Towards Universal Safe Water Access in Rural Tanzania: Moving from Provision to Service Models. Queen Elizabeth Scholarship NextGen Seminar Series. Carleton University. Ottawa, ON [Presentation]

Robbie A. Venis and Onita D. Basu, 2020. Co-application of Silver and Zinc Oxide Nanoparticles for Disinfection in Ceramic Water Filters. University of North Carolina Water & Health Conference. Chapel Hill, NC [Poster]

Robbie A. Venis, Virginia Taylor, and Onita D. Basu, 2018. Challenges and Opportunities for Ceramic Water Filter Implementation in Rural Tanzania. Ontario Water Works Association Annual Conference and Exhibition. Ottawa, ON [Poster]

Non-Peer Reviewed Contributions

Robbie A. Venis, 2021. Rethinking Universal Safe Water Access. Carleton University TEDx Talk Series. Ottawa, ON. [Available]

Robbie A. Venis, 2020. When Local Women Take the Lead: Water Technology Adoption in Rural Tanzania. Carleton University 3-Minute Thesis Competition (2nd Place). Ottawa, ON [Available]

Robbie A. Venis, Onita D. Basu, 2019. Ceramic Water Filters Design and Implementation in Rural Tanzania. Queen Elizabeth Scholarship Marketplace Talk Series. Toronto, ON.

Robbie A. Venis, 2018. Canadian Engineering Talent Helps Develop Water Filters in Africa. CBC Radio, Quirks and Quarks. Ottawa, ON. [Available]

Appendix E Chapter 4 Statistical Tables

Please note that all Tukey's HSD tests use α = 0.05.

Table E1. Tukey's HSD Table for 1 mg/L Ag at different water chemistries

HSD = 0.158414465; df = 139; $n_{treatments}$ = 5							
	0 minutes	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Water Chemistry	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
6.4 High DO - 7.4 Synth	9.5E-18	0.02718	0.0692	0.1533	0.1055	0.1142	0.2152
6.4 High DO - 8.4 Synth	0.0E+00	0.03871	0.1352	0.0926	0.0865	0.1296	0.526
6.4 High DO - 7.4 Raw	0.0E+00	0.04685	0.1321	0.1658	0.0266	0.1445	0.8066
6.4 High DO - 6.4 Low DO	0.0E+00	0.10663	0.1721	0.2667	0.2658	0.416	0.9517
7.4 Synth - 8.4 Synth	9.5E-18	0.01153	0.0661	0.0607	0.019	0.0154	0.3108
7.4 Synth - 7.4 Raw	9.5E-18	0.01967	0.0629	0.0126	0.0789	0.0304	0.5914
7.4 Synth - 6.4 Low DO	9.5E-18	0.07945	0.1029	0.1135	0.1603	0.3018	0.7365
8.4 Synth - 7.4 Raw	0.0E+00	0.00814	0.0032	0.0732	0.0598	0.0149	0.2806
8.4 Synth - 6.4 Low DO	0.0E+00	0.06792	0.0368	0.1742	0.1793	0.2863	0.4257
7.4 Raw - 6.4 Low DO	0	0.05978	0.04	0.1009	0.2392	0.2714	0.1451

Table E2. Tukey's HSD Table for 1 mg/L Zn at different water chemistries

HSD = 0.13219355; df = 139; $n_{treatments}$ = 5							
	0 minutes	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Water Chemistry	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
6.4 High DO - 7.4 Synth	5.6E-18	0.0467	0.1304	0.0676	0.117	0.1446	0.1907
6.4 High DO - 8.4 Synth	0.0E+00	0.0618	0.1084	0.0169	0.0288	0.001	0.1778
6.4 High DO - 7.4 Raw	9.1E-18	0.1719	0.2256	0.2082	0.2381	0.2463	0.3741
6.4 High DO - 6.4 Low DO	0.0E+00	0.14	0.2267	0.1549	0.2429	0.2517	0.3496
7.4 Synth - 8.4 Synth	5.6E-18	0.015	0.022	0.0507	0.0881	0.1456	0.0129
7.4 Synth - 7.4 Raw	3.5E-18	0.1252	0.0953	0.1405	0.1212	0.1017	0.1834
7.4 Synth - 6.4 Low DO	5.6E-18	0.0933	0.0963	0.0873	0.1259	0.1072	0.1589
8.4 Synth - 7.4 Raw	9.1E-18	0.1101	0.1172	0.1913	0.2093	0.2473	0.1963
8.4 Synth - 6.4 Low DO	0.0E+00	0.0783	0.1183	0.138	0.214	0.2528	0.1718
7.4 Raw - 6.4 Low DO	9.107E-18	0.0319	0.0011	0.0533	0.0047	0.0054	0.0245

Table E3. Tukey's HSD Table for 0.67 mg/L Ag and 0.33 mg/L ZnO at different water chemistries

HSD = 0.195174942; df = 139; $n_{treatments}$ = 5							
	0 minutes	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Water Chemistry	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
6.4 High DO - 7.4 Synth	0.0E+00	0.0103	0.147	0.1512	0.1041	0.2919	0.1768
6.4 High DO - 8.4 Synth	6.9E-18	0.0619	0.0437	0.1031	0.1285	0.0908	0.2718
6.4 High DO - 7.4 Raw	0.0E+00	0.0149	0.0014	0.0022	0.0646	0.0414	0.938
6.4 High DO - 6.4 Low DO	0.0E+00	0.0291	0.0579	0.109	0.3893	0.6293	2.2436
7.4 Synth - 8.4 Synth	6.9E-18	0.0723	0.1033	0.0481	0.0244	0.2011	0.095
7.4 Synth - 7.4 Raw	0.0E+00	0.0045	0.1484	0.149	0.1686	0.3334	0.7612
7.4 Synth - 6.4 Low DO	0.0E+00	0.0188	0.2048	0.2602	0.4933	0.9212	2.0668
8.4 Synth - 7.4 Raw	6.9E-18	0.0768	0.0451	0.1009	0.193	0.1323	0.6662
8.4 Synth - 6.4 Low DO	6.9E-18	0.091	0.1015	0.2121	0.5177	0.7201	1.9718
7.4 Raw - 6.4 Low DO	0	0.0142	0.0564	0.1112	0.3247	0.5879	1.3056

Table E4. Tukey's HSD Table for 0.33 mg/L Ag and 0.67 mg/L ZnO at different water chemistries

HSD = 0.138926313; df = 139; $n_{treatments}$ = 5							
	0 minutes	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Water Chemistry	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
6.4 High DO - 7.4 Synth	0.0E+00	0.0608	0.1696	0.1109	0.0896	0.1317	0.511
6.4 High DO - 8.4 Synth	0.0E+00	0.0285	0.0815	0.104	0.1216	0.142	0.6533
6.4 High DO - 7.4 Raw	0.0E+00	0.0619	0.1139	0.1147	0.0045	0.1706	0.597
6.4 High DO - 6.4 Low DO	0.0E+00	0.0582	0.109	0.1708	0.1781	0.3493	1.4413
7.4 Synth - 8.4 Synth	0.0E+00	0.0322	0.0881	0.0068	0.032	0.0103	0.1423
7.4 Synth - 7.4 Raw	0.0E+00	0.0011	0.0557	0.0039	0.0852	0.3023	0.086
7.4 Synth - 6.4 Low DO	0.0E+00	0.0026	0.0606	0.06	0.0885	0.2176	0.9303
8.4 Synth - 7.4 Raw	0.0E+00	0.0334	0.0324	0.0107	0.1171	0.3126	0.0563
8.4 Synth - 6.4 Low DO	0.0E+00	0.0297	0.0275	0.0668	0.0565	0.2073	0.788
7.4 Raw - 6.4 Low DO	0	0.0037	0.0049	0.0561	0.1737	0.5199	0.8443

Table E5. Tukey's HSD Table for 0.14 mg/L Ag and 0.86 mg/L ZnO at different water chemistries

HSD = 0.128472235; df = 139; $n_{treatments}$ = 5							
	0 minutes	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Water Chemistry	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
6.4 High DO - 7.4 Synth	1.3E-17	0.0244	0.0258	0.0122	0.0847	0.1213	0.2764
6.4 High DO - 8.4 Synth	0.0E+00	0.0825	0.0965	0.0894	0.0125	0.0853	0.3593
6.4 High DO - 7.4 Raw	6.9E-18	0.0793	0.0486	0.0155	0.0837	0.259	0.742
6.4 High DO - 6.4 Low DO	0.0E+00	0.0335	0.0275	0.052	0.1676	0.259	0.9125
7.4 Synth - 8.4 Synth	1.3E-17	0.0581	0.0707	0.1016	0.0972	0.2067	0.0829
7.4 Synth - 7.4 Raw	2.0E-17	0.0549	0.0228	0.0277	0.001	0.1376	0.4655
7.4 Synth - 6.4 Low DO	1.3E-17	0.0091	0.0017	0.0398	0.083	0.1376	0.636
8.4 Synth - 7.4 Raw	6.9E-18	0.0032	0.0479	0.0739	0.0962	0.3443	0.3827
8.4 Synth - 6.4 Low DO	0.0E+00	0.049	0.069	0.1414	0.1802	0.3443	0.5532
7.4 Raw - 6.4 Low DO	6.93889E-18	0.0458	0.0211	0.0675	0.084	6E-06	0.1705

Table E6. Tukey's HSD Table for each metal at pH of 6.4, High DO

HSD = 0.194860413; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.10183	0.17505	0.24545	0.25121	0.48437	1.21318
No Metal – 100% ZnO	0.15263	0.23132	0.13939	0.21188	0.27357	0.51817
No Metal – 33% ZnO/67% Ag	0.06909	0.10592	0.15986	0.4889	0.97833	3.04091
No Metal – 67% ZnO/33% Ag	0.08239	0.15717	0.17645	0.27386	0.66043	2.12879
No Metal – 86% ZnO/14% Ag	0.01449	0.00833	0.017	0.1668	0.52096	1.56015
100% Ag – 100% ZnO	0.05081	0.05626	0.10606	0.03933	0.2108	0.69501
100% Ag – 33% ZnO/67% Ag	0.03273	0.06913	0.08559	0.23769	0.49396	1.82773
100% Ag – 67% ZnO/33% Ag	0.01944	0.01788	0.069	0.02266	0.17605	0.91562
100% Ag – 86% ZnO/14% Ag	0.11632	0.18338	0.22845	0.0844	0.03658	0.34697
100% ZnO – 33% ZnO/67% Ag	0.08354	0.12539	0.02047	0.27702	0.70476	2.52275
100% ZnO – 67% ZnO/33% Ag	0.07024	0.07415	0.03706	0.06199	0.38685	1.61063
100% ZnO – 86% ZnO/14% Ag	0.16712	0.23964	0.12239	0.04507	0.24738	1.04199
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.0133	0.05124	0.01659	0.21503	0.31791	0.91212
33% ZnO/67% Ag – 86% ZnO/14% Ag	0.08358	0.11425	0.14286	0.32209	0.45738	1.48076
67% ZnO/33% Ag – 86% ZnO/14% Ag	0.09688	0.1655	0.15945	0.10706	0.13947	0.56864

Table E7. Tukey's HSD Table for each metal at pH of 7.4, High DO

HSD = 0.122574956; df = 167; $n_{treatments}$ = 6							
	0 min	**10 Minutes**	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	9.54E-18	0.023463	0.058821	0.137993	0.271028	0.465502	1.225614
No Metal – 100% ZnO	5.64E-18	0.007798	0.05389	0.117538	0.220248	0.2243	0.55514
No Metal – 33% ZnO/67% Ag	0	0.039346	0.20583	0.356845	0.718313	1.365569	3.091729
No Metal – 67% ZnO/33% Ag	0	0.076475	0.059494	0.111366	0.309564	0.624065	1.845411
No Metal – 86% ZnO/14% Ag	1.3E-17	0.088196	0.029626	0.050638	0.207473	0.494918	1.511366
100% Ag – 100% ZnO	3.9E-18	0.031261	0.004931	0.020455	0.05078	0.241202	0.670474
100% Ag – 33% ZnO/67% Ag	9.54E-18	0.015883	0.147009	0.218851	0.447285	0.900067	1.866115
100% Ag – 67% ZnO/33% Ag	9.54E-18	0.053012	0.118315	0.026627	0.038536	0.158563	0.619798
100% Ag – 86% ZnO/14% Ag	3.47E-18	0.064733	0.088447	0.087355	0.063555	0.029416	0.285752
100% ZnO – 33% ZnO/67% Ag	5.64E-18	0.047144	0.15194	0.239307	0.498066	1.141269	2.536589
100% ZnO – 67% ZnO/33% Ag	5.64E-18	0.084273	0.113384	0.006172	0.089316	0.399765	1.290271
100% ZnO – 86% ZnO/14% Ag	7.37E-18	0.095994	0.083516	0.0669	0.012775	0.270618	0.956226
33% ZnO/67% Ag – 67% ZnO/33% Ag	0	0.037129	0.265324	0.245479	0.408749	0.741504	1.246318
33% ZnO/67% Ag – 86% ZnO/14% Ag	1.3E-17	0.04885	0.235456	0.306207	0.51084	0.870651	1.580364
67% ZnO/33% Ag – 86% ZnO/14% Ag	1.3E-17	0.011721	0.029868	0.060728	0.102091	0.129147	0.334046

Table E8. Tukey's HSD Table for each metal at pH of 8.4, High DO

HSD = 0.142429178; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.07024	0.107401	0.237149	0.25947	0.442216	0.81437
No Metal – 100% ZnO	0.097981	0.190499	0.206747	0.277779	0.362057	0.467549
No Metal – 33% ZnO/67% Ag	0.138136	0.217153	0.34724	0.712107	1.15662	2.896245
No Metal – 67% ZnO/33% Ag	0.061002	0.14323	0.156669	0.24699	0.605891	1.602643
No Metal – 86% ZnO/14% Ag	0.075086	0.155751	0.190685	0.274051	0.693758	1.328027
100% Ag – 100% ZnO	0.027741	0.083098	0.030402	0.018308	0.080158	0.346821
100% Ag – 33% ZnO/67% Ag	0.067895	0.109752	0.110091	0.452637	0.714405	2.081875
100% Ag – 67% ZnO/33% Ag	0.009238	0.035829	0.080481	0.01248	0.163675	0.788273
100% Ag – 86% ZnO/14% Ag	0.004846	0.048351	0.046464	0.014581	0.251542	0.513656
100% ZnO – 33% ZnO/67% Ag	0.040154	0.026654	0.140494	0.434328	0.794563	2.428696
100% ZnO – 67% ZnO/33% Ag	0.036979	0.04727	0.050078	0.030789	0.243833	1.135094
100% ZnO – 86% ZnO/14% Ag	0.022895	0.034748	0.016062	0.003727	0.331701	0.860478
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.077134	0.073923	0.190572	0.465117	0.55073	1.293602
33% ZnO/67% Ag – 86% ZnO/14% Ag	0.06305	0.061401	0.156555	0.438056	0.462862	1.568219
67% ZnO/33% Ag – 86% ZnO/14% Ag	0.014084	0.012522	0.034017	0.027061	0.087867	0.274617

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Table E9. Tukey's HSD Table for each metal in natural water

HSD = 0.14676087; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.040918	0.059902	0.16421	0.346763	0.464508	0.557393
No Metal – 100% ZnO	0.033332	0.022604	0.015762	0.095925	0.151937	0.294898
No Metal – 33% ZnO/67% Ag	0.040161	0.121439	0.246644	0.546528	1.061577	2.25373
No Metal – 67% ZnO/33% Ag	0.006454	0.06023	0.146283	0.391593	0.955698	1.68259
No Metal – 86% ZnO/14% Ag	0.050742	0.057183	0.117087	0.205324	0.386673	0.969013
100% Ag – 100% ZnO	0.07425	0.037298	0.148448	0.250837	0.312571	0.262496
100% Ag – 33% ZnO/67% Ag	0.000757	0.061537	0.082434	0.199765	0.597069	1.696336
100% Ag – 67% ZnO/33% Ag	0.034464	0.000328	0.017927	0.04483	0.49119	1.125196
100% Ag – 86% ZnO/14% Ag	0.009824	0.002719	0.047124	0.141439	0.077835	0.41162
100% ZnO – 33% ZnO/67% Ag	0.073493	0.098835	0.230882	0.450602	0.90964	1.958832
100% ZnO – 67% ZnO/33% Ag	0.039786	0.037626	0.130521	0.295667	0.803761	1.387692
100% ZnO – 86% ZnO/14% Ag	0.084074	0.034579	0.101325	0.109399	0.234736	0.674115
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.033707	0.061209	0.100361	0.154935	0.105879	0.57114
33% ZnO/67% Ag – 86% ZnO/14% Ag	0.010581	0.064256	0.129558	0.341204	0.674904	1.284716
67% ZnO/33% Ag – 86% ZnO/14% Ag	0.044288	0.003047	0.029197	0.186269	0.569025	0.713576

Table E10. Tukey's HSD Table for each metal at pH of 6.4, Low DO

0.075421191; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.012959	0.021336	0.062514	0.109248	0.174539	0.408821
No Metal – 100% ZnO	0.030351	0.022958	0.0683	0.092847	0.127958	0.315902
No Metal – 33% ZnO/67% Ag	0.057756	0.066416	0.134681	0.223483	0.455151	0.944597
No Metal – 67% ZnO/33% Ag	0.04196	0.066532	0.089421	0.21957	0.417277	0.834823
No Metal – 86% ZnO/14% Ag	0.036775	0.037493	0.048832	0.123011	0.368121	0.795014
100% Ag – 100% ZnO	0.017392	0.001622	0.005786	0.016401	0.046581	0.092919
100% Ag – 33% ZnO/67% Ag	0.044797	0.04508	0.072167	0.114234	0.280612	0.535776
100% Ag – 67% ZnO/33% Ag	0.029002	0.045196	0.026907	0.110322	0.242738	0.426002
100% Ag – 86% ZnO/14% Ag	0.023816	0.016157	0.013682	0.013762	0.193582	0.386192
100% ZnO – 33% ZnO/67% Ag	0.027405	0.043458	0.066381	0.130635	0.327193	0.628695
100% ZnO – 67% ZnO/33% Ag	0.01161	0.043574	0.021121	0.126723	0.289319	0.518921
100% ZnO – 86% ZnO/14% Ag	0.006424	0.014534	0.019468	0.030163	0.240163	0.479112
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.015795	0.000116	0.045261	0.003912	0.037874	0.109774
33% ZnO/67% Ag – 86% ZnO/14% Ag	0.020981	0.028923	0.08585	0.100472	0.08703	0.149583
67% ZnO/33% Ag – 86% ZnO/14% Ag	0.005186	0.02904	0.040589	0.09656	0.049156	0.03981

Table E10. Tukey's HSD Table for each metal at pH of 7.4, Low DO

HSD = 0.116671; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.043579	0.071897	0.123227	0.153673	0.244593	0.431716
No Metal – 100% ZnO	0.109417	0.107123	0.106321	0.143879	0.28282	0.446888
No Metal – 33% ZnO/67% Ag	0.073414	0.144134	0.213381	0.447608	0.685515	1.057865
No Metal – 67% ZnO/33% Ag	0.092379	0.090223	0.137549	0.266913	0.503721	0.823817
No Metal – 86% ZnO/14% Ag	0.119144	0.163574	0.286333	0.405308	0.549167	0.905186
100% Ag – 100% ZnO	0.065838	0.035226	0.016906	0.009794	0.038227	0.015172
100% Ag – 33% ZnO/67% Ag	0.029835	0.072237	0.090154	0.293935	0.440921	0.626149
100% Ag – 67% ZnO/33% Ag	0.0488	0.018326	0.014322	0.11324	0.259127	0.392101
100% Ag – 86% ZnO/14% Ag	0.075566	0.091677	0.163106	0.251634	0.304573	0.47347
100% ZnO – 33% ZnO/67% Ag	0.036003	0.037011	0.107059	0.303729	0.402694	0.610977
100% ZnO – 67% ZnO/33% Ag	0.017038	0.016899	0.031227	0.123034	0.2209	0.376929
100% ZnO – 86% ZnO/14% Ag	0.009727	0.056451	0.180012	0.261429	0.266347	0.458298

Table E11. Tukey's HSD Table for each metal at pH of 8.4, Low DO

HSD = 0.078761; df = 167; $n_{treatments}$ = 6						
	10 Minutes	**20 Minutes**	**30 Minutes**	**60 Minutes**	**120 Minutes**	**300 Minutes**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.02179	0.128123	0.166275	0.289129	0.428674	0.588635
No Metal – 100% ZnO	0.036609	0.093918	0.077326	0.105284	0.186156	0.24428
No Metal – 33% ZnO/67% Ag	0.007825	0.053763	0.053643	0.144859	0.237365	0.787705
No Metal – 67% ZnO/33% Ag	0.004006	0.052254	0.075502	0.109192	0.237865	0.64973
No Metal – 86% ZnO/14% Ag	0.004821	0.085506	0.102828	0.209348	0.424838	0.74321
100% Ag – 100% ZnO	0.01482	0.034205	0.088949	0.183845	0.242518	0.344355
100% Ag – 33% ZnO/67% Ag	0.013964	0.074361	0.112632	0.144269	0.191309	0.19907
100% Ag – 67% ZnO/33% Ag	0.017784	0.075869	0.090773	0.179937	0.190809	0.061095
100% Ag – 86% ZnO/14% Ag	0.016969	0.042618	0.063447	0.079781	0.003836	0.154575
100% ZnO – 33% ZnO/67% Ag	0.028784	0.040155	0.023683	0.039575	0.05121	0.543425
100% ZnO – 67% ZnO/33% Ag	0.032604	0.041664	0.001824	0.003908	0.05171	0.40545
100% ZnO – 86% ZnO/14% Ag	0.031788	0.008412	0.025502	0.104064	0.238683	0.49893
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.00382	0.001509	0.021859	0.035667	0.0005	0.137975
33% ZnO/67% Ag – 86% ZnO/14% Ag	0.003004	0.031743	0.049185	0.064488	0.187473	0.044495
67% ZnO/33% Ag – 86% ZnO/14% Ag	0.000815	0.033251	0.027326	0.100156	0.186973	0.09348

Table E12. Tukey's HSD Table for evaluated filter pots

HSD = 0.8611889427; df = 167; $n_{treatments}$ = 6			
	30 Minutes	**60 Minutes**	**24 Hours**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal – 100% Ag	0.219335	0.104809	1.177769
No Metal – 100% ZnO	0.1178	0.077275	0.397349
No Metal – 33% ZnO/67% Ag	0.454819	0.575321	0.927616
No Metal – 67% ZnO/33% Ag	0.27326	0.186157	0.515307
100% Ag – 100% ZnO	0.101535	0.027534	0.780421
100% Ag – 33% ZnO/67% Ag	0.674155	0.68013	0.250154
100% Ag – 67% ZnO/33% Ag	0.053925	0.081348	0.662463
100% ZnO – 33% ZnO/67% Ag	0.572619	0.652596	0.530267
100% ZnO – 67% ZnO/33% Ag	0.15546	0.108882	0.117958
33% ZnO/67% Ag – 67% ZnO/33% Ag	0.72808	0.761478	0.412309

Appendix F - Chapter 5 Statistical Tables

Please note that all Tukey's HSD tests used $\alpha = 0.05$.

Table F1. Tukey's HSD Table for metal elution between evaluated filters

	Ag Elution	**Zn Elution**
Filters	**HSD = 1.52828; df = 39; $n_{treatments} = 5$**	**HSD = 11.5741; df = 39; $n_{treatments} = 5$**
	Abs (μ_1-μ_2)	Abs (μ_1-μ_2)
No Metal - 100% Ag, 0% ZnO	0.95165	1.81175
No Metal - 67% Ag, 33% ZnO	0.54565	4.0563
No Metal - 33% Ag, 67% ZnO	0.353175	1.26055
No Metal - 100% Ag Pot	16.1244	11.53798
100% Ag, 0% ZnO - 67% Ag, 33% ZnO	0.406	5.86805
100% Ag, 0% ZnO - 33% Ag, 67% ZnO	0.598475	0.5512
100% Ag, 0% ZnO - 100% Ag Pot	15.17275	9.726225
67% Ag, 33% ZnO - 33% Ag, 67% ZnO	0.192475	5.31685
67% Ag, 33% ZnO - 100% Ag Pot	15.57875	15.59428
33% Ag, 67% ZnO - 100% Ag Pot	15.77123	10.27743

Table F2. Tukey's HSD Table for bacteria removal by filters at each time of measurement

HSD = 0.82212; df = 299; $n_{treatments}$ = 5					
	30 minutes	**60 minutes**	**24 hours**	**48 hours**	**72 hours**
Filters	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
No Metal - 100% Ag, 0% ZnO	1.061098	0.249311	2.130842	2.106089	2.199496
No Metal - 67% Ag, 33% ZnO	1.056101	0.957521	1.916493	2.398618	2.487483
No Metal - 33% Ag, 67% ZnO	0.210456	0.122616	1.908006	1.161664	0.906063
No Metal - 100% Ag Pot	0.448171	0.62165	2.601855	2.549405	1.974904
100% Ag, 0% ZnO - 67% Ag, 33% ZnO	0.004996	1.206832	0.21435	0.292529	0.287987
100% Ag, 0% ZnO - 33% Ag, 67% ZnO	1.271554	0.126695	0.222836	0.944425	1.293433
100% Ag, 0% ZnO - 100% Ag Pot	0.612927	0.870961	0.471013	0.443316	0.224592
67% Ag, 33% ZnO - 33% Ag, 67% ZnO	1.266558	1.080137	0.008486	1.236954	1.58142
67% Ag, 33% ZnO - 100% Ag Pot	0.60793	0.335871	0.685362	0.150787	0.512579
33% Ag, 67% ZnO - 100% Ag Pot	0.658627	0.744266	0.693849	1.387741	1.068841

Table F3. Tukey's HSD Table for bacteria removal between each time of measurement within each filter type

	No Metal	100% Ag, 0% ZnO	67% Ag, 33% ZnO	33% Ag, 67% ZnO	100% Ag Pot
	HSD = 0.8509; df = 59; $n_{treatments}$ = 5	HSD = 0.6232; df = 59; $n_{treatments}$ = 5	HSD = 0.5724; df = 59; $n_{treatments}$ = 5	HSD = 0.3221; df = 59; $n_{treatments}$ = 5	HSD = 0.4274; df = 59; $n_{treatments}$ = 5
Time	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
0.5 - 1	0.04764	1.262769	0.050941	0.13548	0.221119
0.5 - 24	0.93507	2.004814	1.795461	3.053532	3.088754
0.5 - 48	0.727172	1.772163	2.069688	2.099292	2.828406
0.5 - 72	0.456982	1.59538	1.888364	1.573501	1.983715
1 - 24	0.88743	3.267583	1.846402	2.918052	2.867635
1 - 48	0.679532	3.034932	2.120629	1.963812	2.607287
1 - 72	0.409342	2.858149	1.939304	1.438021	1.762596
24 - 48	0.207898	0.232651	0.274228	0.95424	0.260348
24 - 72	0.478088	0.409434	0.092903	1.480031	1.105038
48 - 72	0.27019	0.176783	0.181325	0.525791	0.84469

Table F4. Tukey's HSD Table for bacteria removal between each metal combination at each time of measurement at initial bacterial concentration of 10^2 CFU/mL

HSD = 0.12418; df = 271; $n_{treatments}$ = 17				
	5 hrs	**24 hrs**	**48 hrs**	**72 hrs**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 0 ppb ZnO	0.057861	0.266465	0.217866	0.093625
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 50 ppb ZnO	0.06545	0.794807	0.911871	0.416177
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 200 ppb ZnO	0.014983	1.541637	1.114153	1.355242
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.197727	3.493577	4.082794	4.454166
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.003069	1.034611	1.311084	1.004955
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.276846	1.193285	1.184191	1.153238
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.07387	1.585531	1.413581	1.643279
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.868537	4.150362	4.270814	4.078368
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.079742	1.626862	1.788789	1.817298
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.108571	2.045412	2.314069	2.091369
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.29512	3.240557	4.334033	4.43561
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.248119	4.109634	4.305344	4.40692
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.51119	2.742361	2.901383	1.898019
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.248613	2.248863	2.315746	1.678878
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.402534	3.935219	4.475982	4.577558
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.506021	4.192793	4.388503	4.490079
1 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 50 ppb ZnO	0.007589	0.528342	0.694005	0.322552
1 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 200 ppb ZnO	0.042878	1.275172	0.896288	1.261617
1 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.255587	3.227112	3.864929	4.360541
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.060929	0.768146	1.093218	0.91133
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.334707	0.92682	0.966325	1.059614
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.016009	1.319066	1.195716	1.549654
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.926397	3.883897	4.052949	3.984743
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.137602	1.360397	1.570923	1.723673
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.166432	1.778947	2.096203	1.997745
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.352981	2.974092	4.116168	4.341985
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.305979	3.843168	4.087478	4.313295
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.56905	2.475896	2.683517	1.804394
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.306473	1.982398	2.09788	1.585254
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.460394	3.668754	4.258116	4.483933
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.563881	3.926328	4.170637	4.396455

1 ppb Ag, 50 ppb ZnO - 1 ppb Ag, 200 ppb ZnO	0.050467	0.74683	0.202283	0.939065
1 ppb Ag, 50 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.263176	2.698769	3.170924	4.037989
1 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.068518	0.239804	0.399213	0.588778
1 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.342296	0.398478	0.27232	0.737061
1 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.00842	0.790724	0.501711	1.227102
1 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.933987	3.355555	3.358944	3.662191
1 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.145192	0.832055	0.876918	1.401121
1 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.174021	1.250604	1.402198	1.675192
1 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.36057	2.44575	3.422163	4.019433
1 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.313569	3.314826	3.393473	3.990743
1 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.57664	1.947553	1.989512	1.481842
1 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.314063	1.454056	1.403875	1.262701
1 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.467984	3.140411	3.564111	4.161381
1 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.571471	3.397986	3.476632	4.073902
1 ppb Ag, 200 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.212709	1.95194	2.968641	3.098924
1 ppb Ag, 200 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.018052	0.507026	0.19693	0.350287
1 ppb Ag, 200 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.291829	0.348352	0.070037	0.202003
1 ppb Ag, 200 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.058887	0.043894	0.299428	0.288037
1 ppb Ag, 200 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.88352	2.608725	3.156661	2.723126
1 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.094725	0.085225	0.674635	0.462056
1 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.123554	0.503775	1.199916	0.736128
1 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.310103	1.69892	3.21988	3.080368
1 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.263102	2.567996	3.19119	3.051678
1 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.526173	1.200724	1.787229	0.542777
1 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.263596	0.707226	1.201593	0.323637
1 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.417517	2.393582	3.361828	3.222316
1 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.521004	2.651156	3.27435	3.134838
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.194658	2.458965	2.771711	3.449211
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.079119	2.300292	2.898604	3.300927
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.271596	1.908045	2.669213	2.810887
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.67081	0.656785	0.18802	0.375798
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.117985	1.866715	2.294006	2.636868
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.089156	1.448165	1.768725	2.362797
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.097394	0.253019	0.251239	0.018556
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.050392	0.616057	0.222549	0.047246
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.313463	0.751216	1.181412	2.556147
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.050886	1.244713	1.767048	2.775288

1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.204807	0.441642	0.393187	0.123392
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.308294	0.699216	0.305709	0.035913
5 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 50 ppb ZnO	0.273777	0.158674	0.126893	0.148284
5 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.076938	0.55092	0.102498	0.638324
5 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.865468	3.11575	2.959731	3.073413
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.076673	0.592251	0.477705	0.812343
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.105502	1.0108	1.002985	1.086415
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.292052	2.205946	3.02295	3.430655
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.24505	3.075022	2.99426	3.401965
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.508121	1.707749	1.590299	0.893064
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.245544	1.214252	1.004662	0.673923
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.399465	2.900607	3.164898	3.572603
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.502952	3.158182	3.077419	3.485124
5 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 200 ppb ZnO	0.350716	0.392246	0.229391	0.49004
5 ppb Ag, 50 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.591691	2.957077	3.086624	2.925129
5 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.197104	0.433577	0.604598	0.664059
5 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.168275	0.852127	1.129878	0.938131
5 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.018274	2.047272	3.149843	3.282371
5 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.971273	2.916348	3.121153	3.253681
5 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.234344	1.549076	1.717192	0.74478
5 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.028233	1.055578	1.131555	0.52564
5 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.125688	2.741934	3.291791	3.424319
5 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.229175	2.999508	3.204312	3.336841
5 ppb Ag, 200 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.942407	2.564831	2.857233	2.435089
5 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.153612	0.041331	0.375207	0.174019
5 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.182441	0.45988	0.900488	0.448091
5 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.36899	1.655026	2.920452	2.792331
5 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.321989	2.524102	2.891762	2.763641
5 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.58506	1.156829	1.487801	0.25474
5 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.322483	0.663332	0.902165	0.0356
5 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.476404	2.349688	3.0624	2.934279
5 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.579891	2.607262	2.974922	2.846801
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.788795	2.5235	2.482026	2.26107
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.759966	2.10495	1.956745	1.986998
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.573417	0.909805	0.063219	0.357242
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.379582	0.040728	0.034529	0.328552

CCC

5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.357347	1.408001	1.369432	2.180349
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.619924	1.901499	1.955068	2.39949
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.466003	0.215143	0.205167	0.49919
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.637484	0.042431	0.117689	0.411711
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.028829	0.41855	0.52528	0.274071
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.215378	1.613695	2.545245	2.618312
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.168377	2.482772	2.516555	2.589622
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.431448	1.115499	1.112594	0.080721
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.168871	0.622001	0.526957	0.13842
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.322792	2.308357	2.687193	2.76026
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.426279	2.565931	2.599714	2.672781
10 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.186549	1.195146	2.019964	2.34424
10 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.139548	2.064222	1.991275	2.31555
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.402619	0.696949	0.587314	0.19335
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.140042	0.203452	0.001677	0.412491
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.293963	1.889807	2.161913	2.486188
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.39745	2.147381	2.074434	2.39871
10 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.952999	0.869076	0.02869	0.02869
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.21607	0.498197	1.432651	2.537591
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.046507	0.991694	2.018287	2.756731
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.107414	0.694661	0.141948	0.141948
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.210901	0.952236	0.05447	0.05447
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.736929	1.367273	1.403961	2.508901
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.999506	1.86077	1.989597	2.728041
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.845585	0.174415	0.170638	0.170638
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.257902	0.08316	0.08316	0.08316
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.262577	0.493497	0.585637	0.219141
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.108656	1.192858	1.574599	2.679539
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.994831	1.450432	1.48712	2.59206
20 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.153921	1.686355	2.160236	2.898679
20 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.257408	1.94393	2.072757	2.811201
20 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	2.103487	0.257574	0.087479	0.087479

DDD

Table F5. Tukey's HSD Table for bacteria removal between each time of measurement for each metal combination at initial bacterial concentration of 10^2 CFU/ml

	0 ppb Ag, 0 ppb ZnO	**1 ppb Ag, 0 ppb ZnO**	**1 ppb Ag, 50 ppb ZnO**	**1 ppb Ag, 200 ppb ZnO**	**1 ppb Ag, 1000 ppb ZnO**	**5 ppb Ag, 0 ppb ZnO**	**5 ppb Ag, 50 ppb ZnO**	**5 ppb Ag, 200 ppb ZnO**	**5 ppb Ag, 1000 ppb ZnO**	**10 ppb Ag, 0 ppb ZnO**	**10 ppb Ag, 50 ppb ZnO**	**10 ppb Ag, 200 ppb ZnO**	**10 ppb Ag, 1000 ppb ZnO**	**20 ppb Ag, 0 ppb ZnO**	**20 ppb Ag, 50 ppb ZnO**	**20 ppb Ag, 200 ppb ZnO**	**20 ppb Ag, 1000 ppb ZnO**
	HSD = 0.0581; df = 15; n = 4	**HSD = 0.0855; df = 15; n = 4**	**HSD = 0.0656; df = 15; n = 4**	**HSD = 0.1980; df = 15; n = 4**	**HSD = 0.1250; df = 15; n = 4**	**HSD = 0.1149; df = 15; n = 4**	**HSD = 0.0995; df = 15; n = 4**	**HSD = 0.1517; df = 15; n = 4**	**HSD = 0.2461; df = 15; n = 4**	**HSD = 0.1265; df = 15; n = 4**	**HSD = 0.1283; df = 15; n = 4**	**HSD = 0.1032; df = 15; n = 4**	**HSD = 0.1683; df = 15; n = 4**	**HSD = 0.1006; df = 15; n = 4**	**HSD = 0.1070; df = 15; n = 4**	**HSD = 0.1689; df = 15; n = 4**	**HSD = 0.1274; df = 15; n = 4**
Times (hrs)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
5 - 24	1.5675	1.2432	0.7072	0.0109	1.7284	0.5359	0.6511	0.0919	1.7143	0.0204	0.3693	1.3779	1.2940	0.6637	0.4328	1.9652	0.1193
5 - 48	1.7632	1.4875	0.7859	0.6341	2.1219	0.4552	0.8559	0.2758	1.6391	0.0542	0.4423	2.2757	1.2940	0.6270	0.3039	2.3102	0.1193
5 - 72	1.8648	1.7133	1.3832	0.4946	2.3917	0.8629	0.9884	0.1476	1.3451	0.1272	0.1180	2.2757	1.2940	0.4779	0.4345	2.3102	0.1193
24 - 48	0.1957	0.2443	0.0786	0.6232	0.3935	0.0808	0.2048	0.3677	0.0753	0.0338	0.0729	0.8978	0.0000	0.0367	0.1288	0.3451	0.0000
24 - 72	0.2973	0.4701	0.6759	0.4837	0.6633	0.3269	0.3373	0.2395	0.3693	0.1069	0.2513	0.8978	0.0000	1.1416	0.8673	0.3451	0.0000
48 - 72	0.1016	0.2258	0.5973	0.1395	0.2698	0.4077	0.1325	0.1281	0.2940	0.0731	0.3243	0.0000	0.0000	1.1049	0.7384	0.0000	0.0000

EEE

Table F6. Tukey's HSD Table for bacteria removal between each metal combination at each time of measurement at initial bacterial concentration of 10^3 CFU/mL

HSD = 0.178544; df = 207; $n_{treatments}$ = 13				
	5 hrs	**24 hrs**	**48 hrs**	**72 hrs**
Metals	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 0 ppb ZnO	0.143814	0.199321	0.380918	0.192232
0 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.313066	2.652013	2.680575	3.010082
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.10882	0.373924	0.203071	0.195324
0 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.521676	2.984259	3.411023	3.573935
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.1267	0.461111	0.261719	0.22185
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.31114	2.650698	4.14415	4.959245
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.342112	4.113707	5.078402	5.612018
0 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	1.002456	4.437902	5.601471	5.883045
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.29099	0.61739	0.721619	0.966174
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.20965	2.191774	3.512136	4.048858
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.384206	4.840337	6.630812	7.273579
0 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.488793	4.753445	6.162336	6.86143
1 ppb Ag, 0 ppb ZnO - 1 ppb Ag, 1000 ppb ZnO	0.169252	2.452692	2.299657	3.202315
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.034993	0.174602	0.177847	0.003092
1 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.377862	2.784937	3.030105	3.766168
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.017114	0.26179	0.119199	0.414082
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.167327	2.451377	3.763232	5.151478
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.198299	3.914386	4.697484	5.80425
1 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.858642	4.238581	5.220554	6.075277
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.147177	0.418069	0.340702	1.158407
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.065836	1.992453	3.131218	4.241091
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.240392	4.641015	6.249895	7.465811
1 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.34498	4.554124	5.781418	7.053662
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 0 ppb ZnO	0.204246	2.278089	2.477504	3.205406
1 ppb Ag, 1000 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.20861	0.332246	0.730448	0.563853
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.186366	2.190901	2.418856	2.788232
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.001926	0.001315	1.463575	1.949163
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.029047	1.461694	2.397827	2.601935
1 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.68939	1.785889	2.920896	2.872963
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.022075	2.034623	1.958956	2.043908
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.103416	0.460238	0.831561	1.038776
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.07114	2.188324	3.950237	4.263497
1 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.175728	2.101432	3.481761	3.851348

5 ppb Ag, 0 ppb ZnO - 5 ppb Ag, 1000 ppb ZnO	0.412855	2.610335	3.207952	3.769259
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.01788	0.087188	0.058648	0.417174
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.20232	2.276775	3.941079	5.154569
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.233292	3.739783	4.875331	5.807342
5 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.893636	4.063978	5.398401	6.078369
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.18217	0.243467	0.518549	1.161499
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.10083	1.817851	3.309065	4.244182
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.275385	4.466413	6.427742	7.468903
5 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.379973	4.379522	5.959266	7.056754
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 0 ppb ZnO	0.394976	2.523147	3.149304	3.352085
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.210535	0.33356	0.733127	1.38531
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.179563	1.129448	1.667379	2.038082
5 ppb Ag, 1000 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.480781	1.453643	2.190449	2.30911
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.230685	2.366868	2.689403	2.607761
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.312026	0.792484	0.101113	0.474923
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.86253	1.856078	3.219789	3.699644
5 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	0.967118	1.769187	2.751313	3.287495
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 50 ppb ZnO	0.184441	2.189587	3.882431	4.737395
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.215413	3.652595	4.816683	5.390168
10 ppb Ag, 0 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.875756	3.97679	5.339753	5.661195
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.164291	0.156279	0.459901	0.744325
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.08295	1.730663	3.250417	3.827008
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.257506	4.379225	6.369094	7.051729
10 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.362094	4.292334	5.900617	6.63958
10 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 200 ppb ZnO	0.030972	1.463009	0.934252	0.652772
10 ppb Ag, 50 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.691316	1.787204	1.457322	0.9238
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.02015	2.033308	3.42253	3.993071
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.101491	0.458924	0.632014	0.910387
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.073065	2.189638	2.486663	2.314334
10 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.177653	2.102747	2.018187	1.902185
10 ppb Ag, 200 ppb ZnO - 10 ppb Ag, 1000 ppb ZnO	0.660344	0.324195	0.52307	0.271027
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.051122	3.496317	4.356782	4.645843
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.132463	1.921932	1.566266	1.56316
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.042093	0.72663	1.552411	1.661561
10 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.146681	0.639738	1.083935	1.249412

GGG

10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 0 ppb ZnO	0.711466	3.820512	4.879852	4.916871
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.792806	2.246128	2.089335	1.834187
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	0.38175	0.402435	1.029341	1.390534
10 ppb Ag, 1000 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	0.486337	0.315543	0.560865	0.978385
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 50 ppb ZnO	0.081341	1.574384	2.790517	3.082684
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.093215	4.222946	5.909193	6.307404
20 ppb Ag, 0 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.197803	4.136055	5.440717	5.895256
20 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 200 ppb ZnO	1.174556	2.648562	3.118676	3.224721
20 ppb Ag, 50 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	1.279144	2.561671	2.6502	2.812572
20 ppb Ag, 200 ppb ZnO - 20 ppb Ag, 1000 ppb ZnO	0.104588	0.086891	0.468476	0.412149

HHH

Table F7. Tukey's HSD Table for bacteria removal between each time of measurement for each metal combination at initial bacterial concentration of 10^3 CFU/mL

	0 ppb Ag, 0 ppb ZnO	**1 ppb Ag, 0 ppb ZnO**	**1 ppb Ag, 1000 ppb ZnO**	**5 ppb Ag, 0 ppb ZnO**	**5 ppb Ag, 1000 ppb ZnO**	**10 ppb Ag, 0 ppb ZnO**	**10 ppb Ag, 50 ppb ZnO**	**10 ppb Ag, 200 ppb ZnO**	**10 ppb Ag, 1000 ppb ZnO**	**20 ppb Ag, 0 ppb ZnO**	**20 ppb Ag, 50 ppb ZnO**	**20 ppb Ag, 200 ppb ZnO**	**20 ppb Ag, 1000 ppb ZnO**
	HSD = 0.4659; df = 15; n = 4	**HSD = 0.1468; df = 15; n = 4**	**HSD = 0.2038; df = 15; n = 4**	**HSD = 0.0860; df = 15; n = 4**	**HSD = 0.1651; df = 15; n = 4**	**HSD = 0.1838; df = 15; n = 4**	**HSD = 0.3154; df = 15; n = 4**	**HSD = 0.5624; df = 15; n = 4**	**HSD = 0.2258; df = 15; n = 4**	**HSD = 0.0907; df = 15; n = 4**	**HSD = 0.0500; df = 15; n = 4**	**HSD = 0.5196; df = 15; n = 4**	**HSD = 0.6218; df = 15; n = 4**
Time	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)	Abs(μ1-μ2)
5 - 24	1.6894	1.6339	0.6496	1.4243	0.7732	1.3550	0.6502	2.0822	1.7461	1.3630	0.2927	1.7667	1.5753
5 - 48	2.8848	2.6477	0.5173	2.7906	0.0045	2.7498	0.9482	1.8515	1.7142	2.4542	0.4177	2.3618	1.7887
5 - 72	3.5276	3.8636	0.8306	3.8317	0.4753	3.4324	1.1205	1.7423	1.3530	2.8524	0.3116	2.3618	1.8451
24 - 48	1.1954	1.0138	1.1669	1.3663	0.7687	1.3948	0.2980	0.2307	0.0319	1.0912	0.1249	0.5951	0.2135
24 - 72	1.8382	2.2297	1.4801	2.4074	1.2485	2.0775	0.4704	0.3399	0.3930	1.4894	0.0189	0.5951	0.2698
48 - 72	0.6428	1.2159	0.3133	1.0412	0.4799	0.6826	0.1723	0.1092	0.3612	0.3982	0.1060	0.0000	0.0563