

# Safe Drinking Water for Gunungkidul

## -Development of a ceramic filtration for point-of-use drinking water treatment in a tropical karst region-

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## List of Publications

This dissertation presents the most important results of my dissertation, which partly have already been published as journal and proceeding manuscripts. Further publications are being prepared.

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## Zusammenfassung

Mit etwa 840.000 Toten jährlich, sind Durchfallerkrankungen die zweithäufigste Todesursache bei Kindern unter 5 Jahren. Dies gilt vor allem für Entwicklungsländer. Da sich diese Erkrankungen meist auf unsicheres Trinkwasser und mangelnde Hygiene zurückführen lassen, könnte ein Großteil dieser Fälle verhindert werden. In Regionen in denen kein sicheres Trinkwasser verfügbar ist, stellt die Wasseraufbereitung im Haushalt eine der vielversprechendsten Möglichkeiten dar, die Gesundheit der Bevölkerung zu verbessern.

Eine dieser Regionen ist der Distrikt Gunungkidul im südlichen Java, Indonesien. Diese Arbeit beschäftigt sich mit der Entwicklung eines lokal hergestellten Keramikfilters zur Haushaltswasseraufbereitung in Gunungkidul.

Solche Keramikfilter werden von der Nichtregierungsorganisation (NGO) *Potters for Peace* beworben und bereits in einigen Entwicklungsländern eingesetzt. In Indonesien werden sie von der sozialen Organisation *Pelita Indonesia* hergestellt und vorwiegend in Westjava verkauft. Diese Silber-behandelten Filter wurden als Basis für die Entwicklung eines, für die Region Gunungkidul optimierten, Filters herangezogen. Da die Datengrundlage über die Leistung der Filter jedoch sehr unvollständig war, wurden zunächst Labor- und Feldversuche durchgeführt, um die Filter bezüglich Morphologie, Zusammensetzung und Leistung umfassend zu charakterisieren.

Basierend auf den gewonnenen Ergebnissen und Erfahrungen wurden potentielle Schwachstellen identifiziert und diese bei der Entwicklung des optimierten Filters berücksichtigt. Dieser Filter sollte zudem unter Verwendung lokal verfügbarer Materialien in einer reproduzierbaren Qualität von den lokalen Töpfern hergestellt werden können. Eine umfassende Analyse der lokal verfügbaren Rohmaterialien und Brennprozesse diente der Entwicklung einer Rezeptur und eines Brennprogramms für die lokale Produktion der Keramikfilter. Neben Siliziumdioxid und Aluminiumoxid konnte ein hoher Anteil an Eisenoxid in den örtlichen Tonmischungen nachgewiesen werden, was typisch für Steingut ist. Dadurch wurde nicht nur die charakteristische rote Farbe hervorgerufen; solche Bestandteile tragen auch zu einer ausreichenden Festigkeit der Keramik bei niedrigen Brenntemperaturen bei.

Außerdem konnten Arsen und Mangan in den lokalen Tönen nachgewiesen werden. Die Analyse der Eluierbarkeit von Inhaltsstoffen zeigte eine anfängliche Auswaschung von



Arsen und Mangan aus der Keramik. Dabei betragen die anfänglichen Konzentrationen 0,001 bis 0,036 mg/L für Arsen (Richtwert nach WHO 0,01 mg/L) und <0,01 bis 0,72 mg/L für Mangan (gesundheitsbezogener Richtwert nach WHO 0,4 mg/L). Die Werte sanken jedoch nach Filtration einiger Liter Wasser auf Werte nahe der oder unter den Grenzwerten, so dass die Auswaschung bei Einhaltung von Rahmenbedingungen kein Gesundheitsrisiko darstellte.

Lokales Sägemehl und Reishülsen wurden als Porosierungsmittel in verschiedenen Konzentrationen getestet. Durch die Beimischung von 20 % Porosierungsmittel konnte die Porosität der Keramik von 31 % auf etwa 44 % erhöht werden. Bei einem Anteil von 40 % Sägemehl oder Reishülsen betrug die Porosität etwa 50 %. Gleichzeitig wurde damit auch der Durchfluss erhöht. Die hydraulische Leitfähigkeit wurde von  $6,8 \cdot 10^{-8}$  m/s auf  $2 \cdot 10^{-7}$  m/s für 20 % Porosierungsmittel und auf  $2 \cdot 10^{-6}$  m/s für 40 % Porosierungsmittel gesteigert.

Die fraktionierte Filtration mit den Filtern, die unter Verwendung von Reishülsen als Porosierungsmittel hergestellt wurden, erreichte eine Logreduktion für Bakterien von 1,8 Logstufen für *E. coli* und 2,4 bis >4,5 Logstufen für *P. aeruginosa*. Die Logreduktion für Phagen betrug bis zu 0,8 bis 1,0 Logstufen. Filter mit Silber-Coating erreichten höhere Logreduktionen mit bis zu 4 bis 5 Logstufen für Bakterien und 1,2 bis 1,9 Logstufen für Phagen. Die hier entwickelten Filter waren somit vergleichbar mit dem untersuchten, kommerziell erhältlichen Filter.

Im Allgemeinen waren die lokal verfügbaren Materialien geeignet für die Produktion der Keramikfilter. Die üblicherweise von lokalen Töpfern hergestellte Tonmischung und die üblichen Brennvorgänge stellen eine gute Basis für die Produktion der Keramikfilter dar. Lokal verfügbares Sägemehl und Reishülsen können als Porosierungsmittel genutzt werden. Um eine gleichbleibende Qualität der Filter zu gewährleisten, sind jedoch technische Umbauten und Optimierungen der lokalen Brennöfen ratsam.

Diese Arbeit schafft eine umfassende Basis für die Einführung einer lokalen Produktion von Keramikfiltern für die Haushaltswasseraufbereitung in Gunungkidul.

## Abstract

With about 840,000 deaths every year, diarrheal disease is the second leading cause of death for children under the age of five, especially in developing countries. As it is mostly related to unsafe water and sanitation, a huge amount of cases could be prevented. Point-of-use water treatment is one of the most promising interventions with a great potential to improve the population's health in regions where safe drinking water is not available.

This work addresses the development of a locally produced ceramic filter for point-of-use water treatment in the district Gunungkidul in Southern Java, Indonesia, which belongs to the regions without safe water supply.

Ceramic water filters are promoted by the non-governmental organization (NGO) *Potters for Peace* and already manufactured in many developing countries. In Indonesia, they are produced by the social organization *Pelita Indonesia* and mainly promoted in western Java. This silver-treated filter type was used as a base for developing an optimized ceramic filter for the use in Gunungkidul. However, as there was little data available on the performance of these filters, thorough analysis of morphology, composition and performance was conducted in laboratory and field trials.

Based on the results and experiences with this filter, potential weaknesses were identified and addressed in the development of an optimized filter that could be produced in a reproducible quality in local potteries with locally available material.

Beginning with an extensive analysis of local raw material for the filter production and local firing processes, a filter composition and firing procedure was developed for the local production of ceramic filters. Apart from silica and alumina, a high proportion of iron oxide was determined in the local clay mixture which is typical for earthenware clays. Besides the characteristic reddish color, iron oxide contributed to sufficient strength of the ceramic even at low firing temperatures. Moreover, manganese and low concentrations of arsenic could be detected in the local clays. Leachability analysis of the filter revealed an initial leaching of this arsenic and manganese from the ceramic. Initial concentrations were varying from 0.001 to 0.036 mg/L for arsenic (*WHO* guideline value 0.01 mg/L) and <0.01 to 0.72 mg/L for manganese (*WHO* health-based guideline value 0.4 mg/L). However, these values decreased after filtration of several liters of

water to values close to or below guideline values, so that leaching of these elements did not pose an actual health risk.

As pore-forming agents, locally available sawdust and rice husks were tested in different concentrations. Porosities increased from 31 % for ceramics without pore-forming agents to 44 % for 20 % pore-forming agents and about 50 % for 40 % pore-forming agents. Simultaneously, flow rate and hydraulic conductivities increased from  $6.8 \times 10^{-8}$  m/s for 0 % pore-forming agents to  $2 \times 10^{-7}$  m/s for 20 % and  $2 \times 10^{-6}$  m/s for 40 % pore-forming agents.

Fractionated filtration with rice husk filters achieved a bacteria log reduction of about 1.8 log steps for *E. coli* and 2.4 to >4.5 log steps for *P. aeruginosa*. For phages, log reduction in fractionated filtration was up to 0.8 to 1.0 log steps. Filters with silver coating achieved higher log reductions of about 4 to 5 log steps for bacteria and 1.2 to 1.9 log steps for phages. These were comparable to log reductions achieved by the commercially available filter.

Generally, the locally available material proved to be suitable for a production of ceramic filters. The traditionally prepared clay mixture and local firing habits give a good base for production of ceramic filters. Addition of locally available sawdust or rice husks is suitable for attaining the porosity needed for filtration. However, technical adjustments and optimizations of the local kilns are advised to assure consistent quality.

In closing, this work gives an extensive base for implementing a local production of ceramic filters for household water treatment in Gunungkidul.

## Abstrak

Dengan jumlah kematian sekitar 840.000 jiwa setiap tahun, penyakit diare menjadi penyebab kematian utama kedua pada anak berusia dibawah lima tahun, khususnya di negara berkembang. Penyakit ini berkaitan erat dengan kondisi air bersih dan sanitasi yang sebenarnya dapat dilakukan pencegahan kejadian kasus diare dalam jumlah yang besar. Pengolahan air pada titik penggunaan (point of use) merupakan salah satu bentuk intervensi yang sangat berpotensi untuk meningkatkan kesehatan masyarakat yang dihidup di daerah dengan kondisi ketersediaan air yang terbatas.

Penelitian ini dimaksudkan untuk mengembangkan filter keramik lokal untuk pengolahan air pada titik penggunaan (point of use) di Kabupaten Gunungkidul, bagian selatan Pulau Jawa yang merupakan daerah dengan kondisi penyediaan air yang terbatas.

Filter keramik untuk pengolahan air dikenalkan oleh sebuah lembaga swadaya masyarakat (LSM/NGO) bernama *Potters for Peace* dan telah diproduksi di berbagai negara berkembang. Di Indonesia, filter keramik diproduksi oleh organisasi sosial bernama *Pelita Indonesia* dan dikenalkan khususnya pada masyarakat Jawa Barat. Filter inilah yang digunakan sebagai dasar pengembangan dan optimalisasi fungsi filter keramik untuk penggunaan di Gunungkidul. Namun, karena terbatasnya data yang tersedia tentang kemampuan filter tersebut, maka dilakukan analisis morfologi, komposisi dan kemampuannya melalui uji coba dilapangan dan laboratorium. Berdasarkan hasil dan ujicoba yang dilakukan terhadap filter tersebut, beberapa potensi kelemahan telah diidentifikasi dan kemudian dilakukan pengembangan dan optimalisasi filter tersebut dengan melibatkan pengrajin gerabah/keramik setempat dengan bahan/material lokal yang tersedia.

Penelitian dimulai dengan melakukan analisis secara menyeluruh terhadap bahan baku lokal dan proses pembakaran untuk memproduksi filter oleh masyarakat setempat. Sebuah resep yang berisi komposisi bahan dan proses pembakaran telah dikembangkan untuk memproduksi filter keramik lokal. Selain silica dan alumina, kandungan senyawa besi (besi oksida) juga ditemukan dalam kadar tinggi pada campuran lempung lokal yang secara umum merupakan sifat umum lempung di bumi. Komponen tersebut tidak hanya menyebabkan warna kemerahan terhadap hasil pembakaran namun juga menambah kekuatan keramik meskipun hanya dibakar pada suhu rendah. Lempung lokal yang digunakan terdeteksi mengandung unsur mangan

dan arsenik konsentrasi rendah. Berdasarkan analisis, filter menunjukkan adanya rembesan unsur arsenik dan mangan dari keramik saat awal penggunaan. Konstrasinya bervariasi dari 0,001 hingga 0,036 mg/l (nilai standar WHO 0,01 mg/l) untuk arsenik dan <0,01 hingga 0,72 mg/l (nilai standar WHO 0,04 mg/l) untuk mangan. Meski demikian, nilai tersebut semakin menurun setelah penyaringan beberapa liter air mendekati dan dibawah nilai standar sehingga unsur tersebut tidak membahayakan kesehatan.

Sebagai bahan pembentuk pori-pori, bahan lokal berupa serbuk gergaji dan sekam padi halus telah diujicoba dengan kadar yang berbeda. Porositas meningkat dari 31% untuk keramik tanpa tambahan bahan pembentuk pori, menjadi 44% dengan penambahan 20 % bahan, dan porositas menjadi 50% dengan penambahan 40% serbuk gergaji atau sekam padi halus.

Pada saat yang sama, kecepatan menyaring/mengalirkan dari filter keramik juga mengalami peningkatan dengan konduktivitas hidraulik sebesar  $6,8 \cdot 10^{-8}$  meter/detik tanpa penambahan bahan (0%), sebesar  $2 \cdot 10^{-7}$  meter/detik untuk penambahan bahan sebesar 20 % dan sebesar  $2 \cdot 10^{-6}$  meter/detik untuk penambahan bahan pembentuk pori-pori sebesar 40 %.

Penyaringan bertingkat menggunakan filter keramik dengan campuran sekam padi halus menunjukkan adanya penurunan bakteri sebesar 98,4 % untuk *E. coli* dan 99,60 hingga >99,9968 % untuk *P. aeruginosa*. Untuk Phages, penurunan untuk penyaringan bertingkat mencapai 84 hingga 90 %. Filter dengan lapisan perak memiliki nilai pengurangan lebih tinggi mencapai 99,99 hingga 99,999 % untuk bakteri dan 93,7 hingga 98,7 % untuk phages. Nilai-nilai tersebut merupakan hasil dari perbandingan terhadap penurunan log yang didapat dari filter keramik yang dijual secara umum.

Secara umum, material lokal yang tersedia menunjukkan kesesuaian untuk produksi filter keramik. Campuran lempung yang dilakukan secara tradisional serta cara pembakaran yang biasa dilakukan merupakan awalan yang cukup bagus untuk memproduksi filter keramik. Penambahan serbuk gergaji atau sekam padi halus yang banyak tersedia di daerah tersebut sangat cocok untuk memperoleh porositas yang dibutuhkan dalam proses filtrasi. Namun demikian, penyesuaian perlu dilakukan untuk tungku pembakaran supaya dapat memastikan kualitas yang memadai dan seragam.

Sebagai penutup, hasil penelitian ini memberikan dasar yang luas bagi implementasi untuk memproduksi filter keramik untuk pengolahan air rumah tangga di Gunungkidul.

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## Abbreviations and Indonesian terms

<i>BMBF</i>	<i>Bundesministerium für Bildung und Forschung</i> ( <i>Federal Ministry of Education and Research, Germany</i> )
BSA	bovine serum albumin
BW	brook water
CFU	colony forming units
DGGE	denaturing gradient gel electrophoresis
<i>DIN</i>	<i>Deutsches Institut für Norm (German Institute for Standardization)</i>
DNA	deoxyribonucleic acid
dNTP	deoxynucleotide
<i>DINKES</i>	regional health authority ( <i>Dinas kesehatan</i> )
<i>DVGW</i>	<i>Deutscher Verein des Gas- und Wasserfaches e. V.</i> ( <i>German Technical and Scientific Association for Gas and Water</i> )
EC	electrical conductivity
<i>E. coli</i>	<i>Escherichia coli</i>
EDTA	ethylenediaminetetraacetic acid
ED XRF	energy dispersive X-ray fluorescence
<i>EN</i>	<i>Europäische Norm (European standard)</i>
<i>Ent. faecium</i>	<i>Enterococcus faecium</i>
ESEM	environmental scanning electron microscope
F	filtrate
<i>FGK</i>	<i>Forschungsinstitut für Anorganische Werkstoffe – Glas/Keramik – GmbH (Research Institute for Glass and Ceramics)</i>
<i>FGK-SOP</i>	<i>Standard operation procedure of FGK</i>
GSE	gaseous secondary electron
HPC	heterotrophic plate count
HWT	household water treatment
ICP-OES	inductively coupled plasma optical emission spectrometry
IDEASS	Innovation for Development and South-South Cooperation
<i>ISO</i>	<i>International Organization for Standardization</i>

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IWRM	Integrated Water Resources Management
<i>NCBI</i>	<i>National Center for Biotechnology Information</i>
NGO	non-governmental organization
NTU	nephelometric turbidity unit
<i>P. aeruginosa</i>	<i>Pseudomonas aeruginosa</i>
PBS	phosphate buffered saline
PCR	polymerase chain reaction
<i>PDAM</i>	local water authority ( <i>Perusahaan Daerah Air Minum</i> )
PE	polyethylene
<i>PPF</i>	<i>Potters for Peace</i>
PFU	plaque forming units
POU	point-of-use
qPCR	quantitative polymerase chain reaction
RAW	raw water
RH (R)	rice husks
RF	river filtrate
rpm	rounds per minute
RW	river water
SD (S)	sawdust
T	tap water
TAE	buffer containing tris base, acetic acid and EDTA
TOC	total organic carbone
TW	German tap water
<i>UNICEF</i>	<i>United Nations Children's Fund</i>
UV	Ultraviolet (light)
<i>WHO</i>	<i>World Health Organization</i>
XRD	X-ray diffraction

# 1 Introduction

## 1.1 Motivation

About one eighth of the world's population does not have access to safe drinking water (<sup>1</sup>WHO/UN-Water, 2010). Though more and more people are connected to piped systems or use other improved water sources, this does not imply that these water sources are safe. For instance about 1.8 billion people use fecally contaminated drinking water sources (WHO, 2014). Moreover, many diseases, such as diarrhea, cholera, typhoid or hepatitis A, are caused by waterborne pathogens (WHO, 2014). Especially in developing countries people suffer from waterborne diseases (WHO, 2008). The second leading cause of death for children under the age of five is diarrheal disease with 840,000 deaths per year (WHO, 2014). Diarrheal disease is mostly related to unsafe water and sanitation (WHO, 2013) and could be prevented by introducing appropriate measures. Although children, elderly people and immuno-compromized people are most vulnerable to these diseases (Brown, 2007) waterborne pathogens pose a general health risk. In rural developing regions, piped water systems are mostly not available or deliver contaminated water. Alternative water sources such as ground water, river water or rain water are often contaminated during transport and storage, if they have not already been contaminated at the source. In these regions, point-of-use (POU) water treatment often is the only practicable way for people to get safe drinking water (UNICEF/WHO, 2012). There are various technologies for POU water treatment such as solar disinfection, boiling or chlorination. A very promising technology is the use of ceramic water filters like the ones promoted by the non-governmental organization (NGO) *Potters for Peace (PFP)*. In contrast to highly sophisticated technologies, this filter poses a sustainable alternative which is relatively easy to handle and largely accepted by the users. This type of filter is already produced in many developing countries and known to significantly reduce fecal bacteria (Roberts, 2003; Brown, 2007; Duke *et al.*, 2009). Since these filters are locally produced, they exhibit varying characteristics. It has also been reported that metals including manganese and arsenic have been leached from some filters (Van Halem, 2006; Tun, 2009; Archer *et al.*, 2011).

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<sup>1</sup> World Health Organization

Thus, intensive studies of local material and local ceramic processing are necessary for the development of a safe and reliable ceramic water filter. The aim of this dissertation was the development of a locally produced ceramic filter for the project region Gunungkidul in Western Java. Therefore, an evaluation of potential raw material and traditional ceramic processing in the region was conducted. Some aspects that needed to be clarified were the analysis of possible arsenic leaching from the ceramic as well as flow rate and reduction of bacteria depending on composition of the raw material. The effect of silver and fractioned filtration was analyzed to develop an optimized design. Moreover, handling and acceptance needed to be examined for local implementation.

## 1.2 The IWRM project Indonesia

The karst region Gunung Sewu in Southern Java (Haryono and Day, 2004) is one of the developing regions in which POU water treatment poses a great opportunity for the population.

In this region, the population is depending on water from the karst aquifer as there is almost no surface water available due to the poor retention capacity of the karst setting. Especially in the dry season, people are suffering from water scarcity although there is a huge amount of water present in an underground river system which functions as a natural drainage to the Indian Ocean (Uhlig, 1980; Haryono and Day, 2004; Tiede *et al.*, 2007).

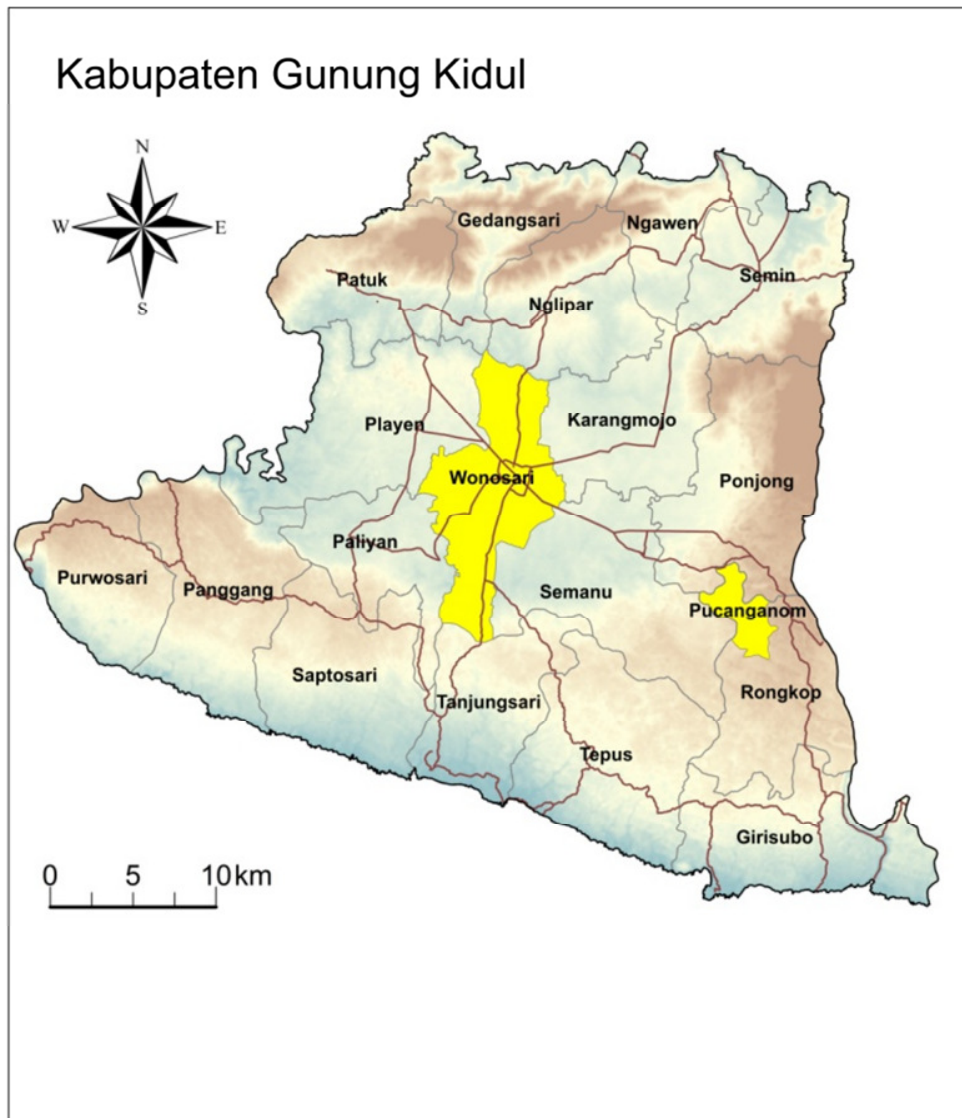
For decades, the Indonesian government has made several attempts to utilize water from the karst aquifer (MacDonald *et al.*, 1984; Lux and Unger 2005). However, none of these attempts has been sustainably successful (Nestmann *et al.*, 2009). Consequently, many households in the project region had to live on less than 10 liters per capita per day in the dry season (Scholz *et al.*, 2004).

Since 2002, a consortium of Indonesian and German partners supported by the <sup>2</sup>BMBF on the German side, has been working on a sustainable solution to make the water from the karst aquifer available for the population. In this context, the world's first underground water extraction plant using hydropower-driven pumping systems was implemented in the cave Gua Bribin. Since the year 2011, the underground water extraction plant has been running 24 hours a day, 7 days a week under the well-trained

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<sup>2</sup> *Bundesministerium für Bildung und Forschung*, Federal Ministry of Education and Research

local operating personnel. It pumps the water continuously into a 220 m elevated reservoir. The innovative underground water supply plant secures the water supply for some 80.000 people by enhancing the available water up to 70 liter per capita per day (Nestmann *et al.*, 2012). Thus, it solved the most urgent problem of the local population. However, after water quantity had been enhanced, further problems needed to be addressed. Karst aquifers are known to be especially vulnerable to pollutions (Kacaroglu, 1999; Goldscheider, 2005; Parise and Gunn, 2007) due to fast and direct infiltration. To make the situation even worse, waste water treatment in the Gunung Sewu region is insufficient or nonexistent. Waste water is partly discharged directly into the ground or collected in nonfunctioning septic tanks (Fach and Fuchs, 2009; Nayono *et al.*, 2010). Considering the low retention capacity for pollutants of the karstic underground, this leads to a high contamination of the karst aquifer with fecal bacteria. Especially in the rainy season, rainwater washes pollutants such as the agricultural waste (e.g. cow dung) and untreated waste water directly into the karst aquifer, which threatens the health of the population. Thus, among others, raw water protection, awareness of the population for environment and hygiene as well as waste water and water treatment needed to be addressed. An integrated approach with appropriate and sustainable technologies and management strategies was needed. These aspects are typically addressed in Integrated Water Resources Management (IWRM) projects. According to *Global Water Partnership (GWP)*, “IWRM is a process which promotes the coordinated development and management of water, land and related resources, in order to maximize the resultant economic and social welfare in an equitable manner without compromising the sustainability of vital ecosystems” (GWP, 2000). Therefore, in 2008 an enlarged consortium of selected Indonesian and German partners from science, industry and government formed to work together in the project “Integrated Water Resources Management in Gunungkidul, Java, Indonesia”. Gunungkidul is a district comprising part of the karst region Gunung Sewu and includes the district capital Wonosari as well as the project’s pilot village Pucanganom (yellow areas in Figure 1). One of the most important aims of the project was the development and implementation of appropriate and sustainable solutions for providing drinking water in sufficient quantity and quality for the population of the district Gunungkidul in Southern Java.



**Figure 1:** Map of the district Gunungkidul. Provided by the Geodetic Institute<sup>3</sup> GIK, KIT. Areas in yellow mark the district capital Wonosari and the pilot village Pucanganom.

For the development of appropriate and sustainable strategies, a comprehensive knowledge of local circumstances and needs is crucial. However, the data available for the project region were very scarce. Therefore, an intensive acquisition of data for hydrological, socio-economic and ecological circumstances as well as physico-chemical and microbiological water analysis was conducted by the project group.

Based on the information and data collected during these studies, the project group was able to develop innovative, appropriate and sustainable technologies for extracting water

<sup>3</sup> Geodätisches Institut, Karlsruhe Institut für Technologie

from the karst aquifer, water distribution, water and waste water treatment and others. Furthermore, strategies and management tools were developed to assure an appropriate and sustainable water management.

The IWRM project Indonesia was characterized by implementation of many strategies and technologies. The micro hydro power plant in the cave Bribin was followed by a pilot plant of a wooden pressure pipeline as an alternative technology. Two field laboratories - one for waste water treatment and one for drinking water treatment - were installed and are now operated by technicians of the hospital in Wonosari. Over the course of the project, the village Pucanganom was chosen as a pilot village, in which several technologies were implemented exemplarily. Water cisterns were built using an innovative composition of concrete which was developed for the project region, biogas reactors were installed and ceramic household water filters were implemented.

To assure a sustainable success of the IWRM project, developments and implementations were combined with a thorough transfer of knowledge (Nestmann *et al.*, 2009). Capacity development included Indonesian-German joint studies, various workshops for different groups of people such as technicians, students, school kids or farmers as well as public relations.

Within the *BMBF* funded IWRM project, many technologies and strategies were developed for sustainable use of the karst aquifer as drinking water source. These are not only appropriate for the project region Gunungkidul, but can be transferred to many other karst regions such as regions in South East Asia or Southern America, in which people suffer from water scarcity while underground rivers carry huge amounts of water into the surrounding oceans.

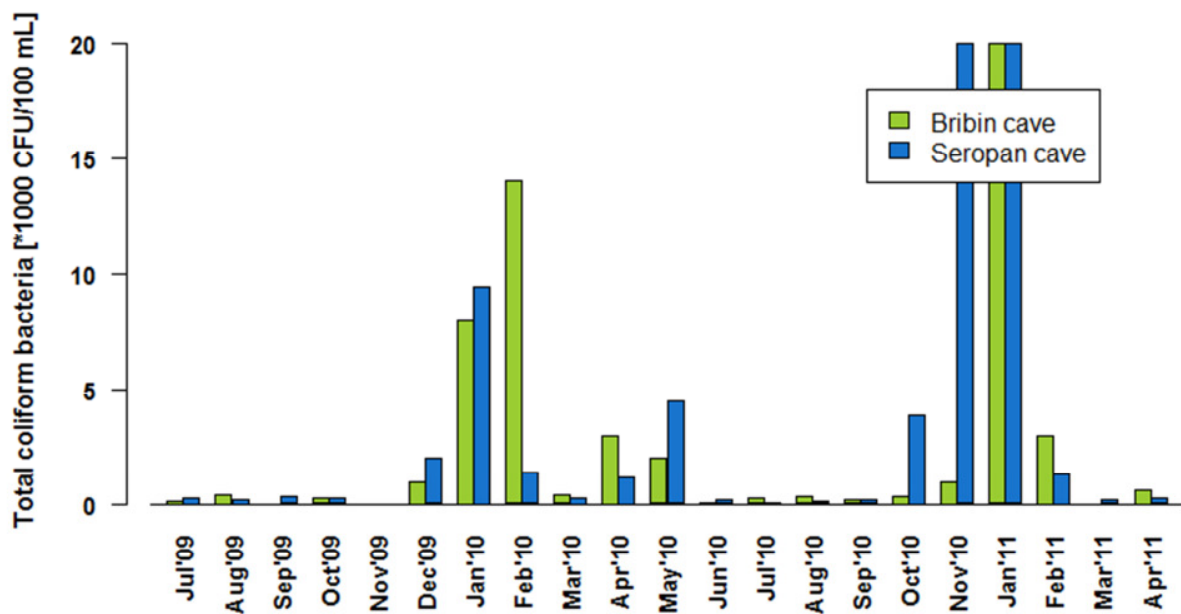
In the following chapters, the water quality monitoring and the drinking water treatment concept that was developed for the project region, are shortly presented. Based on the results obtained in the IWRM project, this dissertation focuses on the development of a POU water treatment with ceramic filters.

### **1.3 Source water quality**

As there were no reliable data available on the water quality in the project region, an intensive water quality monitoring was implemented. For about two years, samples were taken monthly from two cave waters that are used for drinking water purposes. One of



these caves was the Gua Bribin (cave Bribin) whose water is extracted and distributed to the population. The other cave was the cave Seropan which is also used for drinking water purposes. Analysis showed that there were no heavy metals or pesticides in the water. However, turbidity and bacterial contamination were high with extreme peaks in the rainy season. While contamination in the dry season was relatively low, in the rainy season peaks of more than 39,000 CFU/100 mL for coliform bacteria (Matthies *et al.*, 2014) and 9,500 CFU/100 mL for *Escherichia coli* (*E. coli*) could be observed (Figure 2). Drinking water regulations in Indonesia as well as WHO guidelines demand 0 CFU/100 mL for total coliforms and *E. coli* (Menteri Kesehatan Republik Indonesia, 2002; WHO, 2011). Samples never conformed to these guidelines.



**Figure 2:** Coliform bacteria counts in two caves.

Values for both caves in January 2011 and for Seropan cave in November 2010 reached the detection limit of 39,000 CFU/100 mL. In the figure, values are cut at 20,000 CFU/100 mL.

Turbidity peaks in the rainy season reached 500 NTU, while in the dry season, 5 to 10 NTU were determined (Fuchs *et al.*, 2015).

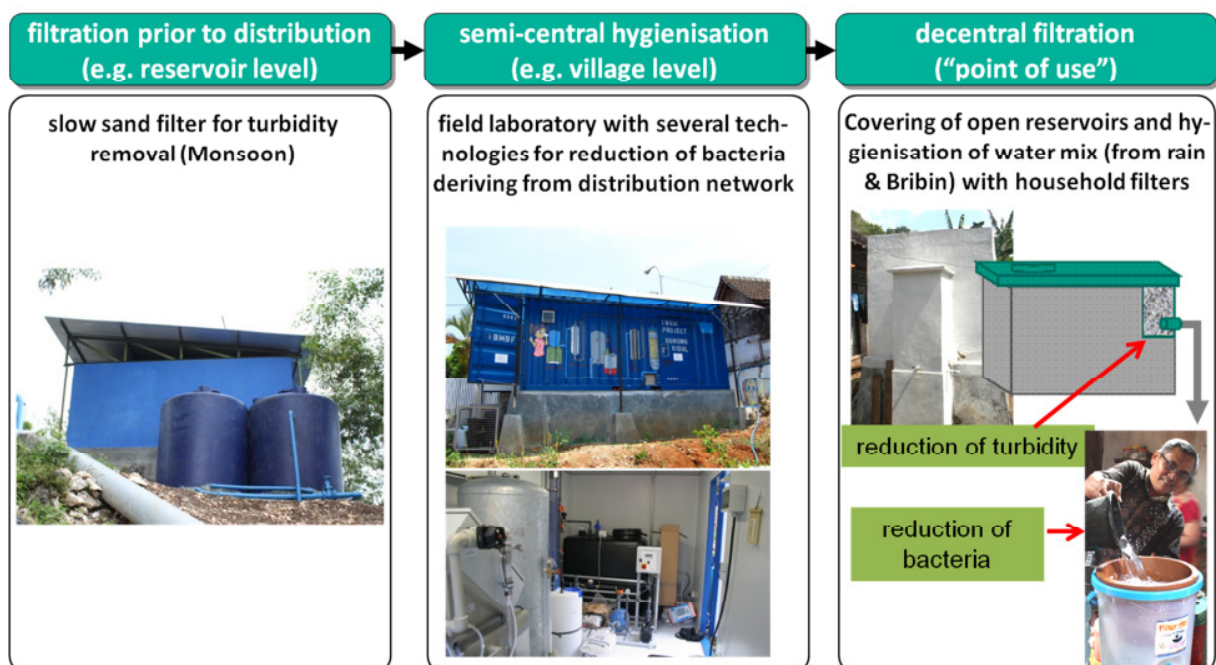
Additionally, the reservoirs along the Bribin distribution system were sampled twice a year. Results showed that the contamination rose along the water distribution system (Matthies *et al.*, 2014). Population analysis revealed that some bacteria species present in the distribution system could not be found in the cave water, suggesting that they derived from the distribution system. Reasons can be found in the dilapidated pipelines

and open storage tanks. The pipelines running above ground are heated by the tropic sun which contributes to an increased growth of bacteria (Matthies *et al.*, 2014). In general, hygienically relevant bacteria that were present in the water of the distribution system (and partly in the cave) comprised *E. coli*, enterococci, pseudomonads including *Pseudomonas aeruginosa* (*P. aruginosa*), *Shigella flexneri*, *Staphylococcus aureus* and *Legionella pneumophila*.

#### 1.4 Developed drinking water treatment concept

To provide drinking water in sufficient quality, an appropriate water treatment concept was needed. Considering the economical and technical circumstances of the region such as the poor infrastructure, low cost solutions with simple technical requirements that could mainly do with locally available material were chosen for drinking water treatment. Based on previous monitoring results, a kind of multi-barrier concept consisting of three steps was developed for water treatment (Matthies *et al.*, 2014).

In Figure 3, an overview about the whole treatment concept and the corresponding implementation in the project region is given. The single treatment steps are addressed in the following paragraphs.



**Figure 3:** Water treatment concept.

**Slow sand filtration.** Monitoring results showed that particles were washed into the karst aquifer leading to extremely high turbidity peaks combined with high numbers of microorganisms in the rainy season (Matthies *et al.*, 2014). Therefore, the first step of the developed water treatment concept is a slow sand filtration that is installed centrally prior to water distribution to remove turbidity and bacteria and thus protect the distribution system. Especially the high peak turbidities in the rainy season are prevented from entering the distribution system. Experiments with local sands, such as beach and lava sand, as well as the planning of the construction of a sand filter were done by the *Department of Sanitary Environmental Engineering* (<sup>4</sup>IWG-SWW, KIT). The construction in Gunungkidul was executed by Indonesian project partners (see Figure 4). More information on the sand filter can be found in Fuchs *et al.*, 2014.



**Figure 4:** Sand filter installed at first reservoir following water extraction.

**Semi-central hygienization.** Whereas central water treatment prior to the water distribution is common practice at any modern waterworks worldwide, the fall-off in water quality during local water distribution demands a hygienization step after the main distribution network (Matthies *et al.*, 2014). This hygienization step is implemented at a semi-central level, but close to the consumer in order to remove bacteria that derive from the dilapidated water distribution system. It could be installed once in every village or at public places where a high amount of water is needed, for instance at schools.

To find the most appropriate technology for this water treatment step, selected technologies were tested under local conditions. Sand filtration was chosen as an

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<sup>4</sup> *Institut für Wasser und Gewässerentwicklung, Bereich Siedlungswasserwirtschaft und Wassergütemirtschaft, Karlsruhe Institut für Technologie*

additional pretreatment to remove turbidity and adhering bacteria. Chlorination and UV disinfection were tested as two standard technologies that are widely spread in industrialized countries. For chlorination, the Indonesian calcium hypochlorite “Kaporit” (tjiwi kimia, Indonesia) was used as it is best available in the project region. These technologies were installed in a shipping container which functions as a field laboratory (see Figure 5) by Berkefeld (ELGA Berkefeld, Veolia Water Solutions and Technologies, Celle, Germany). Finally, an ultra-filtration system (ZIMMERMANN GmbH, Oehringen, Germany) was installed using 50 nm and 200 nm ceramic membranes (atech innovations gmbh, Gladbeck, Germany) which belong to the less expensive ultra-filtration technologies and are relatively stable and comparatively easy to handle.

The four technologies were chosen based on local circumstances, such as availability of basic material as well as adaptation to the poor infrastructure and power supply, to the lack of adequately trained people and to the lack of means for costly water treatment systems.



**Figure 5:** Field laboratory installed at Hospital Wonosari.

The painting explains the water treatment technologies in Indonesian language as the laboratory is situated next to the future public parking of the hospital.

In order to test the technologies and for optimization of the installation, the field laboratory was first installed at a contaminated spring in the Black Forest, Germany. This measure proved to be very important as there were some optimizations needed that would have been very difficult to conduct in Indonesia, such as height adjustment of

the sand filter column or installation of an additional pipe to be able to keep water in a circuit. The technologies showed a good disinfection capacity according to the German drinking water regulations. The optimized field laboratory was then ready to be tested in the project region under local conditions.

Therefore, the container was shipped to Indonesia and installed at the district hospital of Gunungkidul in Wonosari. This location was chosen as it is supplied with water from the local water authority (PDAM) which delivers water that derives from several sources and corresponds to the water in the Bribin system in terms of bacterial load (Matthies *et al.*, 2014). Experiments were conducted with two different water sources to determine the most robust and easiest to handle technology with a good hygienization capacity. Results of the experiments under Indonesian conditions suggested that a combination of sand filtration as a pre-treatment and ultra-filtration with ceramic membranes might be the best solution for this water hygienization step.

Original paper on the field laboratory experiments published in: Matthies K., Mendes de Oliveira M., Obst U. (2014): *Field laboratory for an appropriate water treatment in a tropical karst region*, Water Practice and Technology (doi: 10.2166/wpt.2014.053).

**Third treatment step.** Semi-central solutions for water treatment are feasible for urban areas or at public facilities in which high water amounts are needed and trained personal is available for operation and maintenance. However, in the rural regions where mainly farmers and craftsmen live, no such personal is available and safe drinking water from the piped system still remains decades away.

Moreover, monitoring findings showing that the contamination rises within water distribution and the growing understanding of the local habits during the work in the pilot village Pucanganom emphasized the need of a third treatment step at household level. Currently, several households share one connection to the public water supply in order to save money because water is charged on fixed basis. The piped system is operated intermittently with occasional breakdowns that might last for several weeks. Thus, in order to overcome shortages, people use open tanks to store the water and allow settling of particles. Water from this piped system is untreated, highly contaminated with hygienically relevant bacteria and holds high levels of turbidity in the rainy season. Furthermore, mostly people fed rain water to the turbid and hard tap water in the tanks in

order to get softer and less turbid water. However, due to the fact that these tanks are generally not covered, waste, small animals and other organic matter can enter the water and are decomposed in the tanks. Thus, the water is additionally contaminated and microorganisms are further multiplying in the tropical heat. Analysis showed that bacterial counts were relatively high in these tanks including hygienically relevant bacteria. Moreover, algae as well as potentially toxin producing cyanobacteria could be detected.

Therefore, a water treatment at the household level was urgently needed. Boiling of water has been promoted in the region in the past decades (DINKES, 2011). However, boiling consumes a high amount of energy, and fuel such as wood can be expensive and barely available (Sobsey, 2002). In the project region, 95 % of the local people use biomass (especially wood) as their main or second source of energy (Nayono, 2014). This first has to be collected in the surrounding fields and plantations and carried home which is very time-consuming. Experts estimate that boiling of 1 L of water consumes about 1 kg of wood, which is hardly sustainable (Sobsey, 2002). In addition to the high energy consumption, boiling is quite labor-intensive and produces unhealthy gases. According to the local *Department of Health*, the most frequent illnesses in the region are respiratory diseases (DINKES, 2011) which could also be enhanced by unhealthy cooking and boiling practices in the households. Moreover, the boiling procedure is often performed too short for disinfection or water is just heated and not boiled at all. In interviews, local people revealed another aspect, since with increasing age, people tend to drink unboiled water directly from the tap.

Thus, a more sustainable as well as simple and effective solution for POU water treatment was needed.

## **1.5 Point-of-use drinking water treatment**

Measures for treating water or preventing contamination during storage in the household are called point-of-use (POU) or household water treatment (Brown, 2007).

**Reasons for point-of-use drinking water treatment.** There are various regions worldwide that have no or only insufficient central drinking water treatment. This is especially true for developing countries. Experts estimate that more than 780 million people do not have access to improved water sources. Moreover, many improved water

sources deliver contaminated water (UNICEF/WHO, 2012) and often, water is contaminated during storage or transport (Clasen and Bastable, 2003). For such water sources, POU drinking water treatment might be important to protect public health (UNICEF/WHO, 2009). Furthermore, in many countries and circumstances, POU water treatment is the most cost-effective intervention for preventing waterborne diseases, especially diarrhea (WHO, 2002). There has been a variety of studies proving that POU drinking water treatment has a significant effect in reducing waterborne pathogens (Clasen, 2007; WHO/UNICEF, 2012). Therefore, in the *guidelines for drinking water quality*, the WHO recommends the use of point-of-use water treatment “where piped water systems are not possible and where people rely on source water that may be contaminated or where stored water becomes contaminated because of unhygienic handling during transport or in the home” (WHO, 2011a).

As described in chapter 1.4 there is a piped distribution system in the project region but the water is not treated and the contamination increases along the distribution system. During storage in open tanks at household level, the water is additionally contaminated. Thus, the situation in Gunugkidul exactly corresponds to the cases in which the WHO recommends a point-of-use water treatment.

**Choice of possible technologies.** Candidate technologies can be divided into two basic categories: physical treatment and chemical treatment. Physical methods comprise boiling or heating, filtering, settling and UV radiation whereas chemical methods include coagulation-flocculation, precipitation, ion exchange, chemical disinfection and adsorption (Brown, 2007; WHO, 2011c). In Table 1, a selection of possible technologies for point-of-use water treatment is given with drawbacks and advantages of each technology. UV disinfection and ultrafiltration with membranes are not listed as these technologies require a reliable power supply which is not usually given in every household in the project region.

**Table 1:** Selection of possible technologies for POU water treatment.

Adapted from Lantagne and Clasen (2009), WHO (2011c) and WHO/UNICEF (2012).

Technology	Advantages	Limitations	Log removal
Boiling	No extra material needed, Broadly accepted	No residual against recontamination, Potential burn injuries and respiratory infections, Cost for fuel / time for collecting wood	Bacteria: 6-9 Viruses: 6-9 Protozoa: 6-9
Solar disinfection	Easy operation and maintenance, No operating costs, Minimal chance of recontamination	Pretreatment needed when high turbidity, No visual improvement, Long treatment time depending on sun intensity	Bacteria: 3-5 Viruses: 2-4 Protozoa: 2-4
Ceramic filtration	Easy operation and maintenance, Visual improvement, No operating costs, Local production	No residual against recontamination, Low flow rate, regular cleaning needed, Varying quality of filters, Transport of fragile ceramic	Bacteria: 2-6 Viruses: 1-4 Protozoa: 4-6
Slow sand filtration	Easy operation and maintenance, Visual improvement, No operating costs, longer life, Rel. high flow rate, Local production	No residual against recontamination, Transport of heavy concrete and sand, Periodic cleaning with downtime	Bacteria: 1-3 Viruses: 0.5-2 Protozoa: 2-4
Chlorination	Residual against recontamination, Easy operation and maintenance, Low cost	Removal capacity decreases with turbidity, Taste and odor, Quality control necessary, Chlorination by-products	Bacteria: 3-6 Viruses: 3-6 <sup>5</sup> Protozoa: 3-5
Combined Flocculent/disinfectant powder	Potential reduction of some heavy metals and pesticides, Residual against recontamination, Visual improvement	Requires multiple steps and user support, Most appropriate to deal with high turbidity, Relatively high cost	Bacteria: 7-9 Viruses: 4.5-6 Protozoa: 3-5

<sup>5</sup> For non-*Cryptosporidium*. Log removal for *Cryptosporidium* is 0-1



It is important to mention that every treatment option requires a certain training of the users and probably ongoing support to achieve necessary changes in behavior (Lantagne and Clasen, 2009), and thus a safe and sustainable use of the technologies. However, some technologies require a higher level of understanding or support than others.

Boiling is very effective against all classes of waterborne pathogens but it consumes a lot of energy (Sobsey, 2002) and is almost unviable in the project region (see chapter 1.4). Moreover, boiling with solid fuels, as performed in the project region, leads to increased indoor air pollution and thus may contribute to respiratory diseases. Though solar disinfection is easy and cheap, it is limited to quality and size of available bottles as well as sun intensity (Carey *et al.*, 2011; WHO/UNICEF, 2012). Moreover, solar disinfection has no potential to reduce turbidity which is a main concern of the local people. Thus, neither boiling nor solar disinfection were chosen. Chemical treatment such as chlorination and combined flocculent and disinfection powder require a relatively high understanding of the users. In addition to formation of by-products (Solsona and Méndez, 2003), chlorination with locally available calcium hypochlorite (Kaporit) requires a quality control as adjusting the concentration of free chlorine showed high variations in previous experiments (Matthies *et al.*, 2014). Combined flocculent and disinfection powder is relatively expensive (WHO/UNICEF, 2012) and thus not feasible in the project region. For Gunungkidul, filtration seemed to be the best choice. Both, ceramic and sand filtration remove turbidity and are easily operated and maintained, though both filters need to be cleaned regularly (WHO/UNICEF, 2012).

It was decided to use a gravel filter at the outlet of the optimized covered settling tanks (see Figure 6) as a pre-treatment to remove turbidity and adhering bacteria. An additional optimization of the settling tanks comprised a covering.



**Figure 6:** Left: traditional open tank. Middle and Right: optimized covered tank with gravel filter.

Then, the water was ready to be filtered through a household ceramic filter, basing on the silver-treated filters promoted by *Potters for Peace*, to remove hygienically relevant bacteria. This kind of filter is known to remove pathogens effectively (Roberts, 2003; Brown, 2007; Duke *et al.*, 2009), can be produced locally and is well accepted by the local population. The development of this ceramic filter is addressed in the main part of this dissertation.

**Choice of ceramic filter.** The water used in households of Gunungkidul is composed of piped water and rain water mixed and stored in open settling tanks. People complain mostly about high turbidity and hardness of the water. They are not aware of the role that bacteria play in diarrhea and other illnesses and mostly not even recognize diarrhea as they take it as natural. A very popular misconception is the hardness of the water causing stomachache and kidney stones.

An extensive analysis throughout several dry and rainy seasons showed that the water is contaminated with fecal and other hygienically relevant bacteria, such as *E. coli*, enterococci, *Staphylococcus aureus*, *P. aeruginosa* as well as cyanobacteria. While some bacteria are planctonic, others are particle-associated and come along with high turbidity (Plummer *et al.*, 1987; Dussart-Baptista *et al.*, 2003). Thus, the removal of turbidity is an important parameter for removal of pathogens. According to literature, both (bio)sand filtration and ceramic filtration have a great potential to reduce turbidity and bacteria, whereas ceramic filtration shows better efficiency, especially for bacteria removal (Duke *et al.*, 2011). Reasons can be found in the smaller pore-sizes and the antibacterial effect of the silver coating of the *PFP* filters. The (bio)sand filter develops an optimal bacteria retention with the so-called *schmutzdecke* that first needs to form on

top of the filter. This *schmutzdecke* is a biologically active slime layer which forms on the top 1 to 2 cm of the sand layer, and contains filtered particulate matter and a biofilm consisting of bacteria, algae, protozoa, invertebrates and extracellular polymeric substances (Law *et al.*, 2001; Percival *et al.*, 2000).

Flow rates of ceramic filters are considerably lower than those of sand filters (Duke *et al.*, 2011; WHO/UNICEF, 2012). However, it is still sufficient to supply an average family size and consumption of 2 liters per person per day (Lantagne, 2001). Though ceramic filters need to be checked for quality and require careful transportation because of their fragility (WHO/UNICEF, 2012), they can still be transported on a motor cycle which is the most widely used vehicle in the project region. Thus, ceramic filters are accessible to the whole local population.

As an additional benefit, the local production of ceramic filters has the potential to promote the local economy (WHO/UNICEF, 2012). Considering all these facts, in the project, ceramic filters were chosen as the POU technology.

## **1.6 Ceramic water filters**

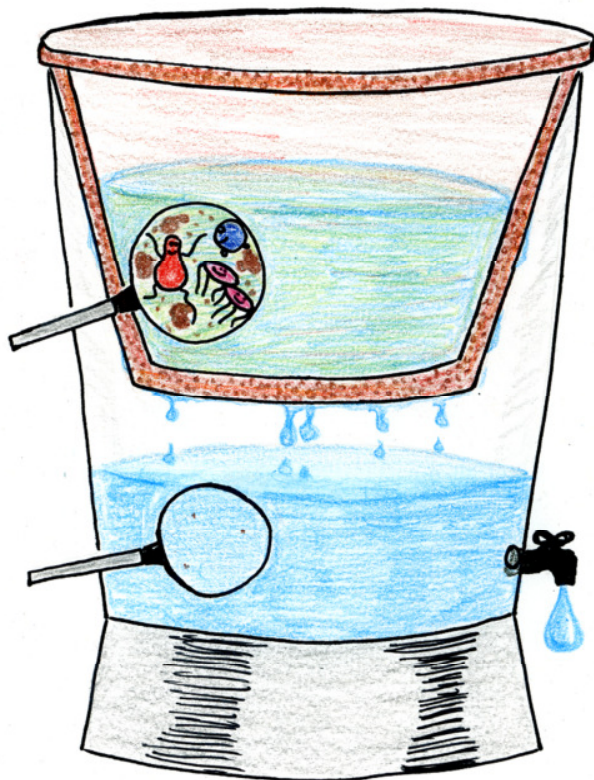
### **1.6.1 Literature review on *PFP* ceramic filters**

Porous ceramic media have already been in use for water treatment in antiquity, and modern records of ceramic household filters can be dated back to the 18<sup>th</sup> century (Baker, 1948; Sobsey, 2002; Brown, 2007). Today, low-cost porous ceramic filters for POU water treatment are typically formed as filter candles or pots (Sobsey, 2002).

One of the most widely used ceramic water filters is the (flower) pot-shaped filter promoted by the NGO *Potters for Peace* (Lantagne *et al.*, 2010; Potters for Peace, 2014) which is also approved by the *WHO* (WHO/UNICEF, 2012). These filters are typically produced by local organizations which get technical support from *Potters for Peace* (*PFP*) for implementation of the filter production (Lantagne *et al.*, 2010). Already manufactured and used in many developing countries such as Cambodia, Ghana and Nicaragua (Van Halem, 2006), the filters are generally made of fired clay. Combustible materials such as rice husks and sawdust are used in the clay mixture to form pores when the material is burned out (Duke *et al.*, 2011). Depending on the available raw material, grog or other material might be added to control porosity (Dies, 2003) or

achieve a more stable product. The filters are typically treated with colloidal silver or silver nitrate solutions to get an additional disinfection effect (Oyanedel-Craver and Smith, 2008; Duke *et al.*, 2011). For instance, the ceramic water purifiers produced in Cambodia which have a capacity of about 10 L are coated with 300 mL of a 0.00215 M silver nitrate solution of which 2/3 are applied on the inside and 1/3 is applied on the outer side of the filter (Brown, 2007).

As shown in Figure 7, users fill the water into the pot from above and allow the water to pass through the porous ceramic walls. The filtered water flows along the outer side to the bottom and drips into a collection receptacle with a tap.



**Figure 7:** Filtration scheme of the *PFP* ceramic filter.

Depending on proper maintenance, the *PFP* filter can have a relatively long lifetime with up to 5 years or even longer (Lantagne, 2001; Campbell, 2005; Brown, 2007). However, it is often recommended to replace the filters after 1 to 2 years (Brown, 2007). The limiting value for the filter lifetime often is the flow rate which is reducing over time but can be restored through scrubbing the filter surface and removing deposits (Lantagne, 2001; Brown, 2007) that block the pores. Therefore, periodical cleaning is necessary.

According to literature, bacteria removal capacities of the *PFP* filters are generally good (Roberts, 2003; Brown, 2007; Duke *et al.*, 2009). However, there are studies showing that not all filters are effective (Campbell, 2005; Lantagne, 2001). For instance, in the study of Enrique Campbell, 2 out of 19 filters (Campbell, 2009) and in the study of Danielle Lantagne even 7 out of 15 filters (Lantagne, 2001) did not show a sufficient removal of *E. coli*. However, it has to be mentioned that these data were acquired in field test, and the reduced removal capacity could be due to an application error and/or lack of cleanliness in the households (Lantagne, 2001). Due to varying raw material and techniques, another known problem with some POU ceramic filters is the leaching of metals (Van Halem, 2006; Tun, 2009; Archer *et al.*, 2011). Thus, as far as feasible under local conditions, an intensive analysis needs to be done for establishing a new filter.

### **1.6.2 *PFP* filter made in Bandung, Indonesia**

As a basis for developing a POU ceramic filter that is optimally adapted to the local conditions in Gunungkidul, an already existing filter produced in Indonesia was analyzed. This kind of *PFP* filter is produced by the social organization *Pelita Indonesia* (*Pelita*) in Bandung, but is mainly promoted in Western Java.

According to information from *Pelita*, the filters are made of local clay and non-clay minerals (such as quartz and feldspars) mixed with sawdust or rice husks depending on availability and price. The ratio of the mixture is adjusted when clay source or combustible material are changed. The mixture is pressed into the pot form, and filters are left in shelves for drying (see appendix) about 2 weeks and even up to 4 weeks in the rainy season (Pelita, 2012).

Then, the filters are fired for about 8 hours with a maximum temperature of about 832 °C (Pelita, 2012). The kiln used at *Pelita* in March 2012 is shown in the appendix. It was damaged by an earthquake which led to 30 % failure during the firing process. Thus, a new optimized kiln was constructed (see appendix).

Filters are checked for their flow rate and if the flow rate is within the limits (1.5 to 2.5 L/h), they are coated with silver nitrate. Otherwise they are discarded and might be used as grog in following mixtures. One filter per batch is checked for removal of coliform bacteria and *E. coli* (Pelita, 2012). However, there were no data available about chemical composition of the filters produced in Bandung and almost no data about the

effect of the filter on the physico-chemical and microbial water quality. To determine microbiological performance and any leaching of metals as well as other effects that might occur, several filters produced by *Pelita* were obtained at the NGO *Yayasan Dian Desa* in Jogjakarta. With these filters, an extensive laboratory analysis in Germany and field studies in Gunungkidul were conducted to determine possible drawbacks in performance, operation and maintenance and to observe the acceptance by local people.

## 1.7 How to evaluate point-of-use water treatment performance

In the WHO guidelines for *Evaluating household water treatment options: Health-based targets and microbiological performance specifications* (WHO, 2011c), advice is given for choice of target bacteria and testing protocols. According to the guideline, test microbes should be selected based on reliable local data on which pathogens have the most significant impact on local waterborne diseases (WHO, 2011c). According to the *Department of Health* in Gunungkidul (DINKES), diarrhea is the most significant waterborne disease in the region (DINKES, 2011; Nayono, 2014). However, it is not further determined which group of pathogens (bacteria, viruses or parasites) is the main source of the diarrhea cases. It can be assumed that eukaryotic parasites are completely removed by the filter due to the small pore size. Therefore, no parasites were tested. Indicator organisms were chosen for bacteria and viruses, based on results obtained in previous studies of the IWRM project.

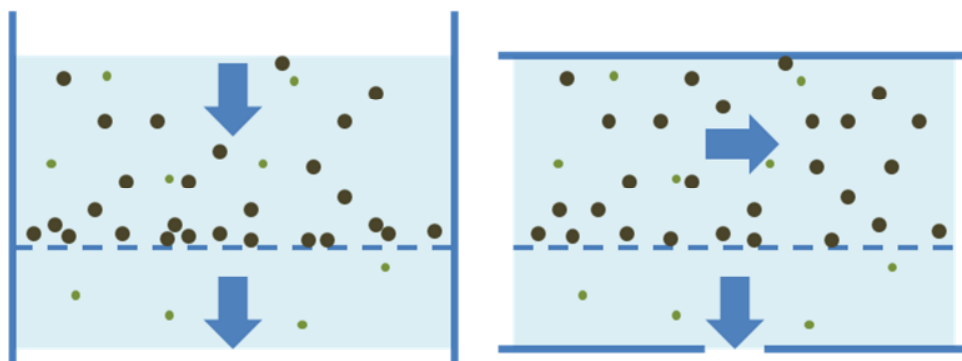
*E. coli* was chosen as it is typically used as an indicator bacterium for fecal contamination and verification of water quality (WHO, 2011a). Additionally, *Pseudomonas aeruginosa* and *Enterococcus faecium* are used as indicator bacteria as all three were previously detected in high amounts in the distributed water (see chapter 1.3). Thus, Gram negative (*E. coli* and *P. aeruginosa*) and Gram positive (*Ent. faecium*) bacteria were tested.

As recommended in literature (WHO, 2011c), the coliphage  $\phi$ X174 was used as surrogate for viruses in the retention experiments as cultivation of viruses was not feasible in the laboratory used for experiments.

## 1.8 Filtration basics

In water treatment, filtration is a basic technology that separates suspended particles from the water. While the particles are kept back on the surface or within the pores of porous membranes or mediums, the fluid filtrate passes through. Filtration can be used to either recover certain valuable substances from the fluid and/or to purify the fluid (Cheremisinoff, 2002). The change in concentration of water components can be due to chemical, physical, biological or combined mechanisms (Karger *et al.*, 2008).

In water treatment, filters are divided in slow filters with filtration rates of 0.05 to 0.3 m/h and rapid filters with filtration rates of up to 30 m/h, though pressure is needed to achieve filtration rates above 15 m/h. Moreover, open and closed systems are differentiated as well as single and multi-layer filters (Karger *et al.*, 2008). Especially when it comes to membrane filtration, filters can be used in a dead-end or in a cross-flow mode (Poirier, 2001) as shown in Figure 8. In contrast to the dead-end mode, in cross-flow filtration, the feed flows tangentially to the filter medium.



**Figure 8:** Schematic of dead-end (left) and cross-flow (right) filtration (Modified after Poirier, 2001).

For both filtration types, the raw water is applied to the feed side of the filter while the filtrate is obtained at the leeward side. It can be distinguished between homogeneous filters made of one single medium and heterogeneous filters made of several filter components. The filtration medium itself is inhomogeneous with pores differing in size, distribution and geometry. Therefore, the flow rate may vary widely within the filter (Cheremisinoff, 2002).

Filter media are characterized by porosity, permeability, tortuosity and connectivity. Porosity describes the hollow fraction of the filter medium, while effective porosity describes the fraction contributing to the flow of a fluid through the medium (Cheremisinoff, 2002).

The most popular equation describing the flow of a fluid through a porous medium is Darcy's law.

$$Q = k_f A \frac{dh}{dx} \quad (1)$$

Q flow rate [m<sup>3</sup>/s]

k<sub>f</sub> hydraulic conductivity

A cross-sectional area (pores and matrix)

h piezometric head

dh/dx hydraulic gradient

With the assumption that the hydraulic gradient along the flow path is linear and the distance between point 1 and point 2 is L, the following equation can be formed:

$$Q = \frac{k_f A (h_1 - h_2)}{L} \quad (2)$$

with

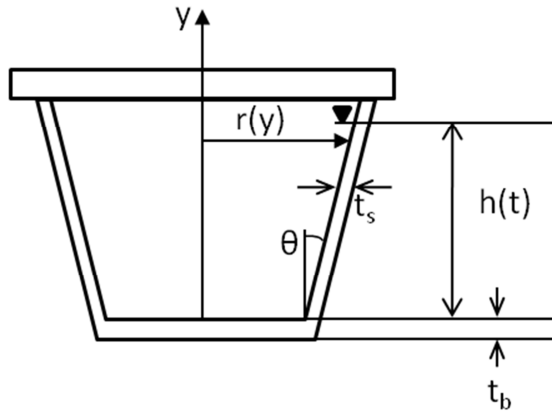
$$k_f = K \frac{\rho}{\mu} g \quad (3)$$

Requirement for Darcy's law is a slow and viscous laminar flow, typically with a Reynolds number below one (Cheremisinoff, 2002).

**Ceramic pot filter.** To describe the flow behavior through ceramic pot filters, a model has been developed by Yakub and Leftwich *et al.* (Leftwich *et al.*, 2009; Yakub *et al.*, 2013).

The basic parameters needed are shown in Figure 9 and include the radius, fluid level (height) and thickness of bottom and side.





**Figure 9:** Schematic depiction of the ceramic pot filter (Modified after Leftwich *et al.*, 2009, Yakub *et al.*, 2013). Variable  $y$  has an upward orientation, radius  $r(y)$  is a function of height, the height of the fluid at a given time is labeled  $h(t)$  and the thickness of bottom and side are labeled  $t_b$  and  $t_s$ .

Based on Darcy's law and with the assumption that thickness of bottom and side are constant, the following equations describe the flow rates through the bottom and the sides of the filter (Schweitzer *et al.*, 2013; Annan *et al.*, 2014).

$$Q_b = \frac{k_f \pi}{t_b} r_0^2 h(t) \quad (4)$$

and

$$Q_s = \frac{2 k_f \pi}{t_s} \int_0^{h(t)} (h - y)(r_0 + y \tan \theta) dy \quad (5)$$

Thus, with integration of (5), the total flow rate is:

$$Q = Q_b + Q_s = k_f \pi \left[ \frac{r_0^2 h(t)}{t_b} + \frac{r_0 h^2(t)}{t_s} + \frac{1 \tan \theta h^3(t)}{3 t_s} \right] \quad (6)$$

This equation is used to compare different filter pots in regard to their hydraulic conductivity.

Filtration processes are separated in cake and depth filtration (Hendricks, 2010). In cake filtration, particles form a cake on the surface of the media which contributes to the filtration process. In contrast, in depth filtration, particles are entrapped in the complex pore structures of the media. Thus, for depth filtration, the filter media normally consists of cartridges or granular media such as sand. The fluid can be forced through the media by gravity, pressure or vacuum as well as centrifugal forces (Cheremisinoff, 2002).

While the fluid passes the filtration media only through the pores, several mechanisms are involved in particle retention (Cheremisinoff, 2002). In general, it can be distinguished between mechanical screening/straining, adsorption, sedimentation and chemical and biological processes (Huisman, 1996; Van Halem, 2006).

Surface straining appears when particles are held back simply because they are larger than the pores. Depth straining describes the retention of particles in the pores when the pore diameter becomes too small during passage of the media (Hutten, 2007). Particle bridging allows retention of particles smaller than the narrowest passage of the filter media (Cheremisinoff, 2002). Further mechanisms of particle capture in depth filtration include inertial impaction, interception, diffusion and electrostatic attraction. Inertial impaction occurs when the particle inertia is high enough for leaving the bending streamline and impacting the filtration media. Diffusion describes the random Brownian motion of very small particles that might cause the particles to leave the streamline and impact the media (Hutten, 2007). Regarding a homogenous spherical particle in a media with cylindrical fibers, interception is described by Lastow and Podgorski as the result of the distance between the particle's center of mass and the media's surface being smaller or equal to the particle radius (Lastow and Podgorski, 1998). Electrostatic attraction can force the particle to leave the streamline when the particle and or the filtration media is electrically or electrostatically charged (Hutten, 2007).

A wide range of particles might be removed from the solid comprising granular materials, fine colloidal particles as well as slimy organic components. Especially colloidal particles and slimy components clog the pores and lead to a fouling of media and decreasing flow rates. To recover the flow rate, the cake on the filter surface or particles within the filter media have to be removed. This can be achieved by scraping, washing or backflushing (Cheremisinoff, 2002).

## 1.9 Ceramic basics

**Ceramics.** Deriving from the Greek term *keramos* (pottery), ceramic refers to products obtained through firing of earthy materials. Ceramics are typically defined as inorganic, non-metallic materials that are produced of clays or other earth minerals as well as chemically produced powders. They are made of compounds between metallic and non-metallic elements and have a crystalline structure (Geiger, 2001).

**Raw material.** According to Guggenheim and Martin, clay is a natural material consisting mainly of fine-grained minerals. While it is normally plastic, clay becomes hard when it is dried or fired. Usually, clay is composed of phyllosilicates (sheet silicate minerals) but might contain further materials leading to plasticity as well as phases including inorganic and organic material not increasing plasticity (Guggenheim and Martin, 1995).

Clay is mainly composed of mineral relics of crystalline source rocks, conversions of sheet silicates or other materials, formations from weathering products as well as organogenic fragments and degradation products (Heim, 1990). Typical relic components are mica, quartz and chlorites while alkali feldspars occur in traces. Moreover, clays normally consist of sheet silicates which can be divided mainly in three mineral groups: smectites, vermiculites and kaolin minerals. Smectites and vermiculites are both fine-grained derivatives from mica (smectites can also be new formations) and are mainly responsible for typical clay characteristics such as ion exchange and plasticity. Kaolin minerals, and especially kaolinite, are formations which can be found with particle sizes above 2  $\mu\text{m}$ . Important non-silicate compounds include carbonate and oxide-hydroxide ferric as well as organogenic substances (Heim, 1990).

Inert materials such as volcanic sands or feldspars as well as lime and gypsum, that are existent in natural clays but might also be added to the clay body, prevent an unwanted deformation during drying and firing. Further additives may be used to adjust the consistency and thus enable certain shaping techniques (Hülseberg, 2014). Organic matter, for instance, may increase the plasticity up to a certain level though further addition of organic matter might lead to a decrease in plasticity (Husein Malkawi *et al.*, 1999).

To gain the necessary porosity of the filters, clay is typically mixed with combustible material (Duke *et al.*, 2011). Under appropriate firing conditions, organic matter may burn completely and lead to an increasing secondary porosity (Maritan *et al.*, 2005).

**Shape.** When sheet minerals come into contact with water, it penetrates between the layers and leads to a plastic mass that can be deformed easily without destroying the bonds between the layers (Hülseberg, 2014).

Depending on the shaping technique, such as casting, twisting/squeezing/pulling as well as pressing, the clay needs to have a low viscosity, high viscosity or dry condition

(Hülseberg, 2014). In case of the ceramic filters, forming on a potter's wheel and axial wet pressing are possible options as they are low cost and easy to perform.

Clay masses for wet pressing typically have a water content of about 12-20 %. The uniaxial pressure leads to a fluid mass and enables a relatively uniform compaction. Moldings need to be dried prior to the firing process (Vogt, 2009).

**Firing/sintering.** Sintering is the part of the firing process of ceramics in which the clay body is compacted and solidified through approaching particles and formation of adhesion bonds. It may be combined with changes in chemical or crystal structure of the ceramic. Moreover, a partial melting phase might occur during sintering (Hülseberg, 2014).

The driving force for the compacting results from the high energy of the particle surfaces. During sintering, the structure and thus the strength and other characteristics of the ceramic are developed. Important parameters are porosity and particle size (Riedel, 2009).

There are different forms of sintering. Solid phase sintering is based on solid body diffusion which is observed in remarkable intensity well above 1000 °C. To achieve a sufficient compaction at lower temperatures, liquid phase sintering is used in which a liquid phase contributes to an improved dissolution and transport of hard materials. Moreover, several forms of reaction sintering and application of pressure to support the compaction are used (Riedel, 2009).

Decomposition and crystallization reactions are influenced by redox conditions, heating rate, and residence time during firing (Maritan *et al.*, 2005).

In the project region, traditional wood-fired kilns are used. Kiln firing comes along with an oxidizing atmosphere, low heating rates and long residence times (Maritan *et al.*, 2005).

## 1.10 Summary / Objective

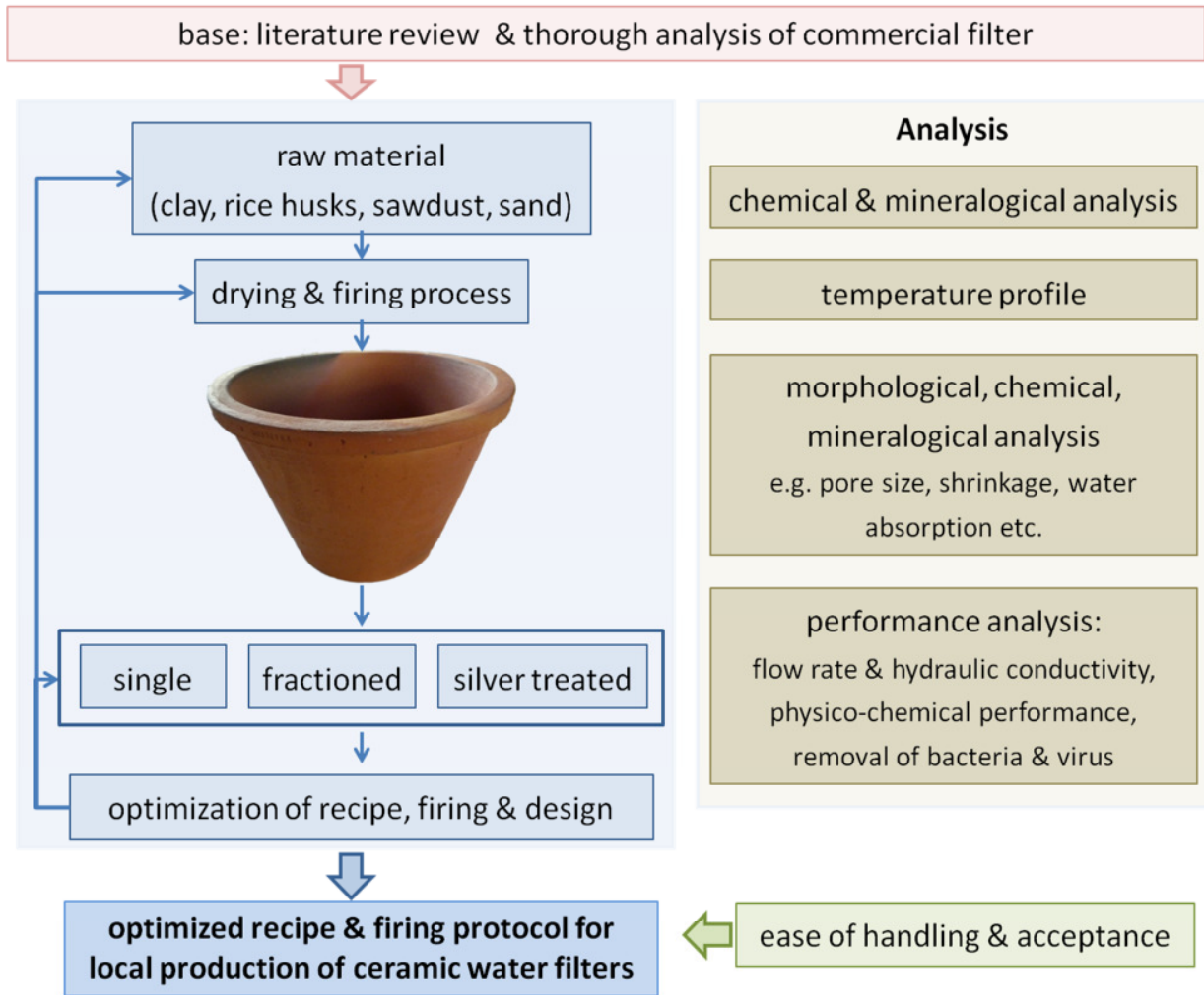
In developing countries, especially in rural regions such as the district Gunungkidul in Southern Java where piped water systems with safe drinking water might be decades away, point-of-use water treatment poses a great opportunity to prevent waterborne diseases and contribute to public health.

Within a German-Indonesian joint IWRM project, a sustainable and appropriate concept for drinking water treatment was developed and implemented. Monitoring results and observations of local circumstances and habits showed that a central water treatment is not feasible and a point-of-use treatment step at household level needs to be included in the concept.

A ceramic filter on the basis of the pot shaped filter promoted by the NGO *Potters for Peace* was chosen for this POU treatment step as it can be locally produced and is known to show good removal capacities for bacteria. However, to make the filter most appropriate for the use in Gunungkidul, field studies needed to be conducted to identify drawbacks that could be addressed in an optimization based on existing filters. The aim of this work was the development of an appropriate composition and firing procedure for the local production of an optimized sustainable ceramic pot filter for the use in Gunungkidul.

Therefore, several aspects needed to be clarified, such as a thorough analysis of an existing filter as a base for optimization, the analysis of local raw material and typical local firing habits, the analysis (e.g. pore size distribution) of ceramic material made of combustible material at varying concentrations, flow rate and hydraulic conductivities of the filter pots, physico-chemical performance, removal efficiency for bacteria and virus, the effect of a fractioned filtration and the effectiveness of a silver treatment.

Figure 10 shows a scheme of the analysis conducted in this work.



**Figure 10:** Scheme of the work conducted in this dissertation.

Combining the results obtained in this work, an appropriate composition and firing procedure for the local production of ceramic filters for the use in Gunungkidul was developed.

## 2 Materials and Methods

Materials that were used repeatedly are given in the following list and are not specified individually within the methods section.

### Equipment:

Autoclave LABOKLAV (SHP Steriltechnik AG, Detzel Schloss/Satuelle, Germany)

Steam pot Varioklav 400DT (H+P Labortechnik GmbH, Oberschleißheim, Germany)

Microwave (Severin Elektrogeräte GmbH, Sundern, Germany)

Elektrophoresis unit Power Pac 200 (Bio-Rad Laboratories GmbH, Munich, Germany)

Lumi-Imager F1<sup>TM</sup> (Roche Diagnostics, Mannheim, Germany)

Centrifuge 5810 R (Eppendorf, Hamburg, Germany)

Centrifuge Biofuge pico (Thermo Fisher Scientific, Dreiech, Germany)

Thermomixer compact (Eppendorf, Hamburg, Germany)

Incubator ICP 600 (Memmert GmbH & Co. KG, Schwabach, Germany)

Incubator KB 400 (Binder GmbH, Tuttlingen, Germany)

Membrane vacuum pump Laboport N820-3FT (KNF Neuberger GmbH, Freiburg, Germany)

Magnetic stirrer MR 3001 (Heidolph, Schwabach, Germany)

pH meter 776 Calimatic (Knick, Berlin, Germany)

Analytical balance Laboratory LC 220 S (Satorius, Göttingen, Germany)

Analytical balance BL 3100 (Satorius, Göttingen, Germany)

Vortex Mixer Stuart (Bibby Sterlin, Staffs, Great Britain)

### Bacteria and Phages:

*Escherichia coli* (DSM 1103, DSMZ GmbH, Braunschweig, Germany)

*Escherichia coli* (DSM 13127 host for phage  $\phi$ X174, DSMZ GmbH, Braunschweig, Germany)

*Escherichia coli* (DSM 5695 host for phage MS2, DSMZ GmbH, Braunschweig, Germany)

*Enterococcus faecium* (DSM 20477, DSMZ GmbH, Braunschweig, Germany)

*Pseudomonas aeruginosa* (ATCC 27853, LGC Standards GmbH, Wesel, Germany)

$\phi$ X174 (DSM 4497, DSMZ GmbH, Braunschweig, Germany)

MS2 (DSM 13767, DSMZ GmbH, Braunschweig, Germany)

**Media and chemicals:**

Acetic acid, CH<sub>3</sub>COOH (VWR International, Darmstadt, Germany)

Bovine serum albumin, BSA (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany)

Calcium chloride, CaCl<sub>2</sub> (Merck KGaA, Darmstadt, Germany)

Cetrimide agar (Merck KgaA, Darmstadt, Germany)

Chromocult® Coliform Agar (Merck KgaA, Darmstadt, Germany)

Chromocult® Enterococci Agar (Merck KgaA, Darmstadt, Germany)

Ethylenediaminetetraacetic acid, EDTA (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany)

GelRed™ nucleic acid stain, 10.000x (BIOTREND Chemikalien GmbH, Cologne, Germany)

Glycerol (Merck KgaA, Darmstadt, Germany)

Hydrochloric acid, HCl 32 % (Merck KgaA, Darmstadt, Germany)

Nitric acid, HNO<sub>3</sub> 65 % (Merck KgaA, Darmstadt, Germany)

Lysogeny broth, LB (Carl Roth GmbH & Co. KG, Karlsruhe, Germany)

Lysozyme (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany)

Magnesium chloride, MgCl<sub>2</sub> (Merck KgaA, Darmstadt, Germany)

Magnesium sulfate heptahydrate, MgSO<sub>4</sub> x 7 H<sub>2</sub>O (Merck KgaA, Darmstadt, Germany)

Sodium chloride, NaCl (Merck KgaA, Darmstadt, Germany)

Nuclease-free water (Life Technologies, Darmstadt, Germany)

37.5:1 acrylamide/bis solution (SERVA Electrophoresis GmbH, Heidelberg, Germany)

R2A agar (Merck KgaA, Darmstadt, Germany)

Sigma 7-9®, Tris base (tris(hydroxymethyl)aminomethane), NH<sub>2</sub>C(CH<sub>2</sub>OH)<sub>3</sub> (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany)

Tris-HCl (Merck KgaA, Darmstadt, Germany)

Triton X-100 (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany)

**Materials:**

Polycarbonate membrane (0.2 µm, Ø 47 mm, Whatman™ GmbH, Lawrence, USA)

Mixed cellulose ester membrane (0.2 µm, Ø 47 mm, Whatman™ GmbH, Lawrence, USA)



Duran bottles and beakers 50 to 5000 mL (DURAN Group GmbH, Wertheim/Main, Germany)

Research® Pipettes 10 µL, 100 µL, 1000 µL, 10 mL (Eppendorf, Hamburg, Germany)

Pipetting tips (Eppendorf, Hamburg, Germany)

Test tubes 0.2 mL, 0.5 mL, 1.5 mL, 2 mL (Eppendorf, Hamburg, Germany)

Centrifuge tubes, 15, 50 mL (Sarstedt, Nümbrecht, Germany)

Nitrile gloves (Ansell Health Care, Brussels, Belgium)

Forceps, reaction vessel supports (VWR International, Darmstadt, Germany)

## 2.1 Repeatedly used methods

In the following paragraphs, methods are described that were used repeatedly throughout the dissertation. The context in which the methods are used is described in chapter 2.2 to 2.3.5.

### 2.1.1 Morphology of ceramic filter pots

**Drying, firing and overall shrinkage.** The clay mass was rehydrated on a plaster board to form a usable mass of which plates were manually formed. Following a drying period of 72 h at room temperature, the plates were further dried in a drying oven for 24 h at 40 °C followed by 24 h at 110 °C. Plates were then fired at the desired temperature in an electrical chamber furnace. Drying, firing and overall shrinkage were determined according to *FGK-SOP*.

Furthermore, **water absorption** was determined following drying at 110 °C.

**Density and open porosity** were measured following *DIN EN 993-1*.

**Incident light microscopy** was used to get a first idea of the ceramic structure depending on concentration of pore-forming agents.

**Pore size distribution** was analyzed using mercury intrusion porosimetry following *DIN 66133*. Therefore, ceramic plates were cut into pieces with a diamond saw and dried at 110 °C. About 0.4 to 1 g of samples were used for the measurement.

## 2.1.2 Mineralogical and chemical analysis of clays and ceramics

**Qualitative mineral phase analysis** was conducted using X-ray diffraction (*DIN EN 13925-1*). Analytically fine samples were first dried at 40 °C for at least 72 hours before powder preparations were produced following the so-called back loading procedure.

**Elementary oxides** were determined with X-ray fluorescence analysis (*DIN 51001*, following *DIN EN ISO 12677*). Samples of about 10 g were coarse-crushed, dried at 110 °C and grinded analytically fine using a vibration disc mill. Fused beads were made with  $\text{Li}_2\text{B}_4\text{O}_7$  in duplicates.

With separate samples, the **loss on ignition** was determined at 1050 °C in duplicates (*FGK-SOP* following *DIN 51081*).

**Elution of metals** was analyzed for a first assessment of possible leaching effects. Therefore, samples were dried at 110 °C and a defined mass was eluted with deionized water for 24 hours. Quantification of selected elements was done with ICP-OES following *DIN EN ISO 11885*.

## 2.1.3 Physico-chemical performance of filter pots

The physico-chemical performance analysis of the filter bought at *Pelita* is described in chapter 2.2.

For the analysis of the physico-chemical performance of the filters developed in this thesis, samples were taken from raw water and filtrate and stored in glass bottles until analysis. Analysis was conducted at the *CIP Chemisches Institut Pforzheim GmbH* according to the German drinking water regulations (DVGW, 2001). In the following list, the parameters are given with the complying standard test procedures.

<b>Parameter</b>	<b>test procedure (standard)</b>
pH	<i>DIN 38404-C5</i>
electrical conductivity at 25 °C	<i>DIN EN 27888 (C8)</i>
turbidity	<i>DIN EN ISO 7027 (C2)</i>
boron	<i>DIN 38405-D17</i>
chromium, total	<i>DIN EN ISO 11885 (E22)</i>
lead	<i>DIN EN ISO 11885 (E22)</i>
copper	<i>DIN EN ISO 11885 (E22)</i>

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nickel	<i>DIN EN ISO 11885 (E22)</i>
iron	<i>DIN EN ISO 11885 (E22)</i>
manganese	<i>DIN EN ISO 11885 (E22)</i>
sodium	<i>DIN EN ISO 11885 (E22)</i>
potassium	<i>DIN EN ISO 11885 (E22)</i>
magnesium	<i>DIN EN ISO 11885 (E22)</i>
calcium	<i>DIN 38406-E3-3 (calculated)</i>
selenium	<i>DIN EN ISO 11885 (E22)</i>
arsenic	<i>DIN EN ISO 11885 (E22)</i>
antimony	<i>DIN EN ISO 11885 (E22)</i>
aluminum	<i>DIN EN ISO 11885 (E22)</i>
cadmium	<i>DIN EN ISO 11885 (E22)</i>
cyanide, total	<i>DIN 38405-D14-1</i>
mercury	<i>DIN EN 1483 paragraph 4 (E12)</i>
nitrite	<i>DIN EN 26777 (D10)</i>
ammonia	<i>DIN 38406-E5-1</i>
fluoride	<i>DIN EN ISO 10304-1</i>
nitrate	<i>DIN EN ISO 10304-1</i>
chloride	<i>DIN EN ISO 10304-1</i>
sulfate	<i>DIN EN ISO 10304-1</i>
color (absorption at 436 nm)	<i>DIN EN ISO 7887 (C1)</i>
total organic carbon (TOC)	<i>DIN EN 1484 (H3)</i>
oxidizability (expressed in O <sub>2</sub> )	<i>DIN EN ISO 8467 (H5)</i>
total hardness (calcium carbonate)	complexometric titration
total hardness	complexometric titration
carbonate hardness	complexometric titration
acid capacity at pH 4.3	<i>DIN 38409-H7</i>
ortho-phosphate	<i>DIN EN 1189 (D11-3)</i>
calcite solubility (in CaCO <sub>3</sub> )	<i>DIN 38404-C10-R3</i>
absorption at 254 nm	<i>DIN 38404-C3</i>

#### 2.1.4 Microbiological performance analysis of filter pots

**Bacteria removal.** To detect the effectiveness of the filters for bacteria removal, non-sterile tap water was spiked with *Escherichia coli* (DSM 1103), *Pseudomonas aeruginosa* (ATCC 27853) and *Enterococcus faecium* (DSM 20477). Bacteria were cultivated in liquid broth (12.5 g/L LB Broth) at 37 °C for 17 h in a shaking incubator at 160 rpm. Cultures were then centrifuged at 4,000 rpm (Eppendorf centrifuge 5810 R) for 10 min, resuspended in tap water and then mixed with the desired amount of tap water and applied to the filter.

**Colony forming units.** Culture technique with species selective agar was used to detect the bacteria in raw water and filtered water samples. *P. aeruginosa* was detected using Cetrimide agar, *E. coli* using Chromocult<sup>®</sup> Coliform Agar and *Ent. faecium* using Chromocult<sup>®</sup> Enterococci Agar. After incubation at 37 °C for 24 to 48 hours, colony forming units (CFU) were counted.

The **heterotrophic plate count** (HPC) was determined adapted after the German drinking water regulations (DVGW, 2001). Therefore, samples were diluted in 1:10 steps in PBS buffer (137 mM NaCl, 2.7 mM KCl, 0.82 mM Na<sub>2</sub>HPO<sub>4</sub>, 1.5 mM KH<sub>2</sub>PO<sub>4</sub>; pH 7.4). In quadruplicates, 1 mL of each dilution was poured in plates together with molten R2A agar (at 45 °C) and mixed carefully. After solidification, the plates were incubated in duplicates at 22 °C and 37 °C for 72 h and colony forming units were determined. R2A agar was chosen as it is known for its high productivity (Massa *et al.*, 1998).

**Bacteriophage removal.** As recommended in literature (WHO, 2011c), the coliphages  $\phi$ X174 (DSM No. 4497) and partially MS2 (DSM No. 13767) were used as surrogates for viruses in the retention experiments. Therefore, tap water was spiked with a  $\phi$ X174 or MS2 phage suspension to attain a phage concentration of about 10<sup>2</sup>-10<sup>3</sup> PFU/L. This raw water was then applied to the filter.

To detect the bacteriophages in the raw water and filtrate, they were concentrated as follows: a mixed cellulose ester membrane was first washed with 25 mL 185 mM MgCl<sub>2</sub> solution (5 min incubation, filtration at 550 mbar). The samples were mixed with 50 mM MgCl<sub>2</sub> and filtered through the membrane at 550 mbar. The membrane was transferred into a petri dish containing 3 mL elution buffer (3 % meat extract, 3 % Tween<sup>®</sup> 80, 0.75 M NaCl, pH 9) and phages were eluted from the membrane by shaking at 60 rpm

for 60 min. Serial dilutions of the obtained phage solution were prepared in elution buffer for the plaque assay. Plaque assay was performed in duplicates with the appropriate host *E. coli* cultures (DSM 13127 for  $\phi$ X174 and DSM 5695 for MS2). Bottom agar was made of Chromocult<sup>®</sup> Coliform Agar and top agar consisted of 25 g/L Nutrient broth NB 10.1 g/L agar agar, 1.23 g/L MgSO<sub>4</sub> and 0.1 mol/L CaCl<sub>2</sub>. 100  $\mu$ L sample and 100  $\mu$ L host culture (OD<sub>600</sub> ca. 0.5) were mixed with 5 mL molten top agar (45 °C), poured onto the bottom agar and allowed to solidify. After incubation at 37 °C for 24 hours, plaque forming units (PFU) were determined.

### 2.1.5 Molecular biological analysis of raw and filtered water

In addition to the microbiological analysis, culture-independent molecular biological methods were used to analyze the performance of the filters regarding bacteria and phage reduction efficiency. The great benefit of these technologies is the possibility to detect microorganisms that are not easily cultivatable in the laboratory and to overcome selection during cultivation.

For deoxyribonucleic acid (DNA) analysis of bacteria in the samples, 100 mL of the respective sample was filtered through a 0.2  $\mu$ m polycarbonate membrane filter using vacuum filtration. The filter was stored in a 1.5 mL test tube at 4 °C until further analysis. DNA analysis of samples for phage  $\phi$ X174 was done directly from the phage solution obtained from the concentration step in 2.1.4.

**DNA extraction.** DNA was isolated from the samples using the QIAamp DNA Mini Kit (Qiagen GmbH, Hilden, Germany) according to the manufacturer's instructions with the following modifications. Protocol D for isolation of genomic DNA from Gram-positive bacteria was applied, using enzyme solution (20 mg/mL lysozyme, 20 mM Tris-HCl, pH 8.0, 2 mM EDTA, 1.2 % Triton<sup>®</sup>). For final DNA elution, 50  $\mu$ L of PCR grade water was added to the column, incubated at room temperature for 5 min, followed by 5 min centrifugation at 6,000 g. The elution step was repeated once.

**Polymerase chain reaction (PCR).** For DNA amplification, several primer pairs were used depending on the target bacteria and phages. All primers were purchased from Sigma-Aldrich Chemie GmbH (Taufkirchen, Germany). Details for used primer pairs and related temperature profiles are given in Table 2. In general, a 25  $\mu$ L reaction volume with 1 x PCR buffer Y (20 mM Tris-HCl (pH 8.55), 16 mM (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, 0.01 % Tween 20

und 2 mM MgCl<sub>2</sub>) (peqlab Biotechnologie GmbH, Erlangen, Germany) was used, containing 5 µL of template DNA, 25 µg bovine serum albumin (BSA), 200 µM of each deoxynucleotide (dNTP mix, Applied Biosystems, Darmstadt, Germany), 0.4 µM of each primer (Sigma-Aldrich Chemie GmbH (Taufkirchen, Germany), 0.65 U Hot Taq DNA polymerase (peqlab Biotechnologie GmbH, Erlangen, Germany). Along with the samples, a negative control using nuclease-free water and a positive control using DNA of a reference strain were amplified. Amplification was done with a GeneAmp PCR System 9700 (Applied Biosystems, Darmstadt, Germany). Each temperature profile started with a polymerase activation step of 30 s at 94 °C followed by a degradation step at 94 °C, an annealing step at a primer specific temperature (see Table 2) and an elongation step at 72 °C which were repeated for 30-35 cycles. After a final extension step for 7 min (10 min for  $\phi$ X174 primers) at 72 °C, the PCR products were cooled down to 4 °C to stop the reaction. PCR products were stored at -20 °C until further analysis.

Amplification products were checked with gel electrophoresis. Small PCR products (<100 bp) for *E. coli*, enterococci and *P. aeruginosa* were loaded on a 20 % polyacrylamide gel (with subsequent staining in 1x GelRed), while bigger fragments were loaded on a 1 % agarose gel. The gels were examined under UV illumination at 600 nm and a picture was taken.

**Quantitative polymerase chain reaction (qPCR).** 20 µL reaction volumes contained 1 µL of template, 0.25 µM of each primer and 10 µL KAPA™ SYBR® FAST ABI Prism® qPCR reaction mix (peqlab Biotechnologie GmbH, Erlangen, Germany). These reaction volumes were analyzed in triplicates using the CFX96 Touch™ Real-Time PCR Detection System (Bio-Rad Laboratories GmbH, Munich, Germany). Sterile water was used as no template control (NTC). The temperature profile comprised 2 min at 95 °C and 40 cycles of 15 s at 95 °C and 30 s at the primer-specific annealing temperature (AT). Results were analyzed with the Bio-Rad CFX Manager 3.1 (Bio-Rad Laboratories GmbH, Munich, Germany). Log reduction was calculated using the  $\Delta$ C<sub>q</sub> values between raw water samples and specific sample filtrate with  $\Delta$ C<sub>q</sub>=3.32 being 1 log. Primer pairs for *E. coli* (ECOuidAF/ R), enterococci (Ecst784F/ Enc854R) and *P. aeruginosa* (ecfXRT-F/ R) were obtained from Sigma-Aldrich Chemie GmbH (Taufkirchen, Germany).

**Table 2:** Primers with respective PCR product size, sequence and amplification temperature profile.  
AT=annealing temperature

Primers	Target organism	Sequence (5'-3')	Temperature	Product size	Literature
27 f	Bacteria, 16S rDNA	AGAGTTTGATCMTGGCTCAG	94 °C, 30 s	490 bp	Emitiazi <i>et al.</i> , 2004
517 r		ATTACCGCGGCTGCTGG	55 °C, 30 s  x35 72 °C, 60 s		
ECOuidAF	<i>E. coli</i> , uidA gene	GTGTGATATCTACCCGCTTCGC	94 °C, 30 s	87 bp	Frahm and Obst, 2003
ECOuidAR		AGAACGGTTTGTGGTTAATCAGGA	54 °C, 30 s  x30 72 °C, 30 s qPCR AT = 54 °C		
Ecst784F	Enterococcus sp., 23S rDNA	AGAAATTCCAAACGAACTTG	94 °C, 30 s	92 bp	Frahm <i>et al.</i> , 1998
Enc854R		CAGTGCTCTACCTCCATCATT	60 °C, 90 s  x35 72 °C, 90 s qPCR AT = 60 °C		
Pa23F	<i>P.aeruginosa</i> , 23S rDNA	TCCAAGTTTAAGGTGGTAGGCTG	94 °C, 30 s	93 bp	Volkman <i>et al.</i> , 2007
Pa23Rb		ACCACTTCGTCATCTAAAAGACGAC	54 °C, 30 s  x30 72 °C, 30 s		
ecfXRT-F	<i>P.aeruginosa</i> , ecfX gene	AGCGTTCGTCCTGCACAAGT	qPCR with	81 bp	Clifford <i>et al.</i> , 2012
ecfXRT-R		TCCACCATGCTCAGGGAGAT	AT = 56 °C		
PhiX174-F	φX174, ssDNA genome	GCTTGCGTTTATGGTACG	94 °C, 30 s	1500 bp	Broyles <i>et al.</i> , 2002
PhiX174-R		ATACGAAGGCGCATAACG	54 °C, 60 s  x35 72 °C, 60 s		

**Denaturing gradient gel electrophoresis (DGGE).** To detect shifts in the bacterial population due to filtration, DGGE was conducted according to Muyzer *et al.* (Muyzer *et al.*, 1993), using the forward primer GC-27f (see Table 2) with a GC-clamp (5'-CGC CCG CCG CGC CCC GCG CCC GTC CCG CCG CCG CCC CCG CCC C-3') at the 5' end and reverse primer 517R targeting the eubacterial 16S ribosomal DNA. About 7-18  $\mu\text{L}$  of each PCR product were loaded on a 7.5 % (w/v) polyacrylamide gel containing 40-70 % denaturing agents formamide (VWR International, Darmstadt, Germany) and urea (MP Biomedicals, Santa Ana, CA, USA). TAE electrophoresis buffer (40 mM tris base, 20 mM acetic acid and 1 mM EDTA at pH 8.5) was used. Gels were run at 70 V and 56  $^{\circ}\text{C}$  for about 16-17 h using the DGGE Dcode<sup>TM</sup> universal mutation detection system (Bio-Rad Laboratories GmbH, Munich, Germany). After 15 min staining in TAE buffer (pH 8.0) containing 5 % GelRed, the gels were examined under UV illumination at 600 nm and a picture was taken. Single bands were cut from the gels and transferred into 50  $\mu\text{L}$  of nuclease-free water, incubated for at least 20 h at 4  $^{\circ}\text{C}$  and finally reamplified with the eubacterial primer set 27f/517r for subsequent sequencing.

**Sequencing.** 5  $\mu\text{L}$  of PCR products of single bands were mixed with 5  $\mu\text{L}$  of 5 mM reverse primer 517r and sent to the LIGHTrun<sup>TM</sup> sequencing service (GATC Biotech, Constance, Germany) for sequence analysis. The obtained DNA sequences were blasted at the U.S. *National Center for Biotechnology Information NCBI* data base (<http://blast.ncbi.nlm.nih.gov/Blast.cgi>) for matching organisms.

## 2.2 Methods for analyzing an existing filter as a basis for optimization

The existing concept of the *PF*P filters was used as a basis for developing an optimized ceramic filter. Therefore, morphology, composition and performance of the already existing filter type produced by *Pelita* were analyzed to learn about the functioning of the filter and recognize major drawbacks. The filter for these experiments was bought in January 2012 in Indonesia and brought to Germany where it was analyzed at the *Institute of Functional Interfaces* at the *KIT*. Moreover, several samples of filters that had not been coated with silver nitrate were obtained from *Pelita* in March 2012.



**Morphology.** To determine the structure of the filters, several filter fragments that were not coated with silver nitrate were analyzed. Scanning electron micrographs were obtained of all sides of one fragment with an environmental scanning electron microscope (ESEM, (Philips XL-30 ESEM-FEG, FEI Company, Hillsboro, USA)) using a gaseous secondary (GSE) detector system. The acceleration voltage was 20 kV and chamber pressure was about 120 Pa (0.9 Torr). The pore size of the filter is an important factor influencing bacteria removal capacity, so pore size analysis of two fragments was conducted at the <sup>6</sup>*Research Institute of Glass and Ceramics (FGK)* using mercury porosimetry, to get an idea of the pore size distribution.

**Mineralogical and chemical composition.** Analysis for chemical composition and qualitative phase analysis of the ceramic were carried out with bulk powder from an uncoated fragment using X-ray diffraction analysis (XRD). The diffractometer (Bruker/Siemens D5000, Bruker-AXS, Karlsruhe, Germany) uses CuK $\alpha$  radiation and is equipped with a graphite secondary monochromator. The elemental composition of the filter was analyzed using energy dispersive X-ray fluorescence ED XRF (Epsilon 5, PANalytical, EA Almelo, Netherlands). Bulk powder samples were sealed with a 6  $\mu$ m Mylar film in spectro cups. A tungsten X-ray tube was used as the radiation source, with a Ge-detector for detection and quantification.

**Physico-chemical performance.** To analyze the filter's chemical performance, a series of experiments with tap water (TW) and natural river water (RW) was conducted. These were done with the original silver nitrate-coated filter obtained from *Pelita*. First, 3 L of tap water were applied to the filter and the filtrate (F1) was kept for further analysis. This experiment was repeated with 3 L for the second filtrate (F2) and 5 times with 5.5 L each time, for F3 to F7. This series was performed mainly to determine any leaching effects from the filter, and the experiment was repeated subsequently with 6 L of RW (River Rhine at km 362.07, Germany, January 8<sup>th</sup> 2013) to analyze any removal capacity of the filter. For all experiments, the following parameters were determined directly from the raw water and the filtrate: pH (pH/EC/TDS meter, HANNA Instruments Deutschland GmbH, Kehl am Rhein, Germany), electrical conductivity (WTW LF 330, Weilheim, Germany), nitrate and hardness (both: Hach Lange, Düsseldorf, Germany). To

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<sup>6</sup> *Forschungsinstitut für Anorganische Werkstoffe – Glas/Keramik – GmbH*

determine any metal leaching, the concentration of metal ions was analyzed in both raw water and filtrate. The focus was on metals that were detected previously by chemical composition analysis of the filter (see chapter 3.1): As, Ba, Cd, Cr, Mn, Pb and Zn, as well as Ag because of the silver coating. Therefore, 100 mL of each filtrate was applied to a cation exchanger (Cation exchanger I, Merck KGaA, Darmstadt, Germany). Ions were then eluted with 5 mL 4 M nitric acid. The concentrations of metal ions were determined using inductively coupled plasma optical emission spectrometry (ICP-OES, (Optima 8300 DV ICP-OES Spectrometer, PerkinElmer, Rodgau, Germany)). Analysis for arsenic was executed by the *CIP Chemisches Institut Pforzheim GmbH* according to *DIN EN ISO 11885, E22*.

For the experiments with natural river water, turbidity (WTW Turb 355 IR, Weilheim, Germany) and total organic carbon (TOC) concentrations (Multi N/C® 2000, Analytik Jena AG, Jena, Germany) were additionally determined. Turbidity was determined directly for both raw and filtered water. For the TOC analysis, the water was first passed through a 0.45 µm filter (Puradisc 25 TF; Whatman, Lawrence, USA) to remove bigger particles that would disturb the measurement.

To evaluate the suitability of the ceramic filter for use at household level, the flow rate of the filtrate was determined during the experiments.

**Microbiological Performance.** To detect the effectiveness of bacteria removal, tap water was spiked with *E. coli*, *P. aeruginosa* and *Ent. faecium*. Bacteria were cultivated as previously described and then mixed with 7 L of tap water of which 5 L were applied to the filter. Raw water and filtrate were examined as described in chapter 2.1.4 to detect the log reduction achieved by filtration.

Moreover, the effectiveness in phage removal was detected. Therefore, 6 L of tap water were spiked with a φX174 or MS2 phage suspension to attain a phage concentration of about  $10^2$ - $10^3$  PFU/L. 5.5 L of this raw water were then applied to the filter. The remaining 0.5 L of raw water and 1 L of filtrate were concentrated for phage detection.

Additionally, the tap water was mixed with phages and 0.08 g/L loess (Heilerde 1, Luvos®, Germany) to determine any possible effect of turbidity on phage removal efficiency.

Experiments were conducted in duplicate, and analysis of the raw water and filtrates was again done in duplicate.

**Field experiments.** Water samples were taken from field experiments in Indonesia and analyzed at the *CIP Chemisches Institut Pforzheim GmbH*. Raw water, the filtrate of an old filter that had been in daily use for 2 years and the filtrate of a new filter were analyzed.

## **2.3 Analysis needed for the development of an optimized filter to be locally produced**

The optimized filter was supposed to be produced locally and with locally available material. Therefore, appropriate firing procedures and compositions needed to be developed. As a prerequisite, local raw material and local firing processes needed to be analyzed.

### **2.3.1 Analysis of local raw material**

Following the *PFP* protocol, ceramic pot filters are normally produced of local clay, water and additional supporting material depending on the local clay sources as well as combustible pore-forming agents such as rice husks or sawdust (Potters for Peace, 2014).

As the process as well as the product quality very much depend on the available raw material, clays, non-clay minerals (such as lava sand), sawdust and rice husks were analyzed.

The first step was a thorough interview of local potters regarding the clay sources they use, the composition of their typical clay mixture and the usual processing. Several samples of single raw materials and the locally produced clay mixture as well as samples of sawdust and rice husks were analyzed in the laboratories of the *FGK*. An overview is given in Table 3.

**Table 3:** Samples taken in Indonesia and analyzed in the *FGK*.

Sample name	date	origin
Soil	8/2013	Local (Kasongan)
Soil 2	3/2014	Kulon Progo
Red clay 1	8/2013	Mangunan, Bantul
Red clay 2	3/2014	Imogiri
Yellow clay 1	8/2013	Godean
Yellow clay 2	3/2014	Borobodur
Brown clay	3/2014	Bangun Jiwo
Lava sand	8/2013	Progo river
Mixture	2/2013	Kasongan
Sawdust	6/2013	Sewon, Bantul
Rice husks	6/2013	Sewon, Bantul

A **qualitative mineral phase analysis** of the samples was conducted using X-ray diffraction as previously described. Moreover, elementary oxides and loss on ignition was determined.

**Grain size analysis** of samples soil, red clay 1, yellow clay 1 and lava sand was performed using wet screening (*DIN 66165-1/-2*). In an overhead shaker, the samples were dispersed in 0.1 %  $\text{Na}_4\text{P}_2\text{O}_7$  for at least 12 hours. The solutions were then screened through 0.125 to 0.063 mm sieves and flushed with water until the wash float was clear. The passed fractions were recovered.

As the clay samples had a great fraction below 0.063 mm, this fraction was additionally analyzed using light-scattering technique (*DIN EN 725-5*). A sample of the fraction was dispersed in the analysis unit and measured under constant stirring without use of ultrasound.

Finally, **elution of metals** was analyzed for a first assessment of possible leaching effects.

### 2.3.2 Analysis of local firing process

A possible local production of ceramic pot filters is depending not only on the local raw material but also on drying conditions, available kilns and firing skills. Thus, an analysis

of traditional local firing processes was conducted. As no data were available on the actual temperature profile during firing, several firing processes were analyzed.

To observe the temperature profile during the whole firing process, a temperature probe with a hand-held data logger (HH506RA, Newport Electronics GmbH, Deckenpfronn, Germany) was used. It was fixed at different positions during each firing process.

The differences in temperature throughout the kiln were determined using process temperature control rings (RTC AQS 660-1100 °C, M.E.Schupp® Industriekeramik GmbH & Co. KG, Aachen, Germany) that were distributed in the kiln (front and back, top and bottom). These rings shrink during the firing process correlated to the maximum temperature. After firing, the rings were collected and their diameter was measured with digital caliper gauges (DIN 862, MIB Messzeuge GmbH, Spangenberg, Germany). Thus, the maximum temperature achieved at each tested spot and variations throughout the kiln were determined.

### **2.3.3 Development of an appropriate composition**

According to *PPF*, the ceramic filters are typically made of local clays and combustible material in a ratio of 60:40 (Potters for Peace, 2014). For the experiments with ceramic pots, the local clay mixture produced by local potters was shipped to Germany. It composed of several clay sources as well as lava sand and soil which had a high proportion of sand (see chapters 2.3.1 and 3.3.1). In literature, sawdust, corn flour, wheat flour and rice husks can be found as combustible material (Dies, 2003). Due to availability in the project region, sawdust and rice husks were chosen for trial. Both were obtained in June 2013 in Sewon, Bantul, Indonesia which is close to Kasongan where the potteries are located.

After shipping, analysis was done by the *FGK* in Germany. In a first set of experiments, the local clay mixture was mixed with sawdust and rice husks in volume-ratios 80:20 and 60:40. Therefore, the moist local clay mixture was first coarse-crushed manually and then dried in a drying oven at 40 °C for more than 72 h. Afterwards, the clay mass was crushed with a jaw crusher (pulverisette 1 model II, Fritsch GmbH, Idar-Oberstein, Germany), grade 3 followed by crushing at grade 1. The grounded material was then mixed with tap water in a hand-held blender to form a usable mass. Parts of this mass were additionally mixed with sawdust or rice husks at 20 % and 40 % each (see Table

4). The samples were stored over night in polyethylene (PE) bags to guarantee an even spread of the moist and an optimal swelling of the clayey components before they were rehydrated again to form a usable mass of which plates were manually formed.

Drying, firing and overall shrinkage were determined as described in chapter 2.1.2 with a firing temperature of 830 °C in an electrical chamber furnace. This temperature was chosen in accordance with the firing protocol used by *Pelita*.

Density, open porosity, pore size distribution as well as water absorption were determined as previously described.

**Table 4:** List of samples prepared at *FGK* from local material.

Sample name	description
0 % plate	Local clay mixture without further addition of combustible material
20 % S plate	Local clay mixture with 20 % sawdust
40 % S plate	Local clay mixture with 40 % sawdust
20 % R plate	Local clay mixture with 20 % rice husks
40 % R plate	Local clay mixture with 40 % rice husks

To examine possible problems that might occur when the mixture of clay and combustible material is processed locally, rice husks were obtained in Sewon, Bantul and brought to the local clay processing. Workers were told to mix their traditional clay mixture with the rice husks in ratios 4 + 1 (20 % rice husks) and 3 + 2 (40 % rice husks). The mixing process was observed and mixtures (0 %, 20 % and 40 % rice husks) were shipped to the *FGK* in order to be compared with previous *FGK* samples.

Drying, firing and overall shrinkage were analyzed in 8 replicates. Density, open porosity and water absorption were determined as previously described. Moreover, pots were formed from the mixtures (0 % pot, 20 % R pot, 40 % R pot) and flow rates were determined as before.

**Silver.** Silver nitrate (Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany) was used for coating. In a first experiment a filter pot was submerged in a 1.26 g/L silver nitrate solution (in distilled water) for 1 min. After 3 min of “sitting”, the mass of soaked silver solution was determined.

### 2.3.4 Analysis needed for development of an appropriate firing procedure

Details on temperatures used within the firing process for ceramic filters range from 800 (Pelita, 2014) to 870 (Brown, 2007) or 890 °C (IDEASS, 2014). In order to develop an appropriate firing procedure for Gunungkidul, experiments with local raw material needed to be conducted.

**Local material fired in Germany.** Therefore, samples of traditionally used clay mixtures produced from local potters were taken in February 2013, June 2013, August 2013, March 2014 and October 2014 and shipped to Germany for further analysis.

**Melting behavior** of the clay mixture was analyzed using hot stage microscopy (*DIN 51730*). Out of two samples of the traditional local clay mixture a portion of 10 g each was homogenized through manual kneading. After drying at 100 °C, the samples were grounded with a mortar. Cylinders with a height and diameter of 3 mm were formed and analyzed. The samples were heated up to 1500 °C with a heating rate of 5 K/min.

**Firing color, shrinkage, density and water absorption** were determined depending on the firing temperature. Out of the samples of the traditional clay mixture, 12 plates were manually formed and dried at room temperature for 48 h before further drying in the drying oven as described in chapter 2.1.2.

After measuring the drying shrinkage, the samples were fired in an electrically heated gradient kiln at 800, 850, 900, 950, 1000 and 1050 °C for 60 min. For each temperature, the samples were tested in duplicate. Moreover, density, open porosity and water absorption were measured.

**Extension shrinkage analysis** (*DIN 51045*). A 100 g sample of the traditional clay mixture was homogenized through manual kneading and a plate was manually formed. Before analysis, the plate was dried at 40 °C and ground with abrasive paper to a dimension of 25 mm x 5 mm x 5 mm.

**Elementary oxides and loss on ignition** were determined as previously described.

**Local firing.** In a second series of experiments, 10 pots were formed in a local pottery from the traditional local clay mixture using a potter's wheel. The pots were dried for at least 3 to 5 days and then fired at two different temperature profiles. For the first firing

event, potters were asked to reach about 800 °C in the kiln, while the second firing event was following their routine firing habits. Maximum temperatures were determined using process temperature control rings. Moreover, the temperature profile of the first firing process (with higher temperature) was recorded with a temperature probe and data logger as described previously in chapter 2.3.2.

Firing color and stability were recorded before pots were broken into pieces which were shipped to the *FGK* and further analyzed. Density, open porosity and water absorption were determined as previously described.

### 2.3.5 Filter performance and development of an optimized design

**Hydraulic conductivity.** In order to evaluate the flow rates of the filter pots containing different nature and quantity of combustible material, the flow rates were determined for different water heights in the filter. Therefore, pots were first soaked with water and then filled completely. The water was allowed to pass the filter and the water height was noted every 5 (in the beginning) to 15 minutes. The experiment was repeated up to 9 times. The hydraulic conductivity was calculated according to the equation given in chapter 1.8.

**Physico-chemical performance of filter pots.** In order to get first impressions on filter performance of the local clay mixture containing sawdust or rice husks as pore-forming agents, three pots containing 40 % sawdust and three pots containing 40 % rice husks were formed. The clay mixture, sawdust and rice husks were obtained in Indonesia. Mixing, firing experiments and analysis were conducted in Germany. The pots were fired at 850 °C with a heating rate of 2.3 K/min and a retention time of 30 min.

Tap water was applied to a filter pot containing 40 % of sawdust in a series of 1 L each. A physico-chemical analysis (see chapter 2.1.3) of the filtrates was done to determine possible changes in the parameters due to filtration.

**Effect of different concentrations of rice husks.** Subsequently, three filter pots were formed from clay mixtures produced in Indonesia containing 0, 20 and 40 % of rice husks. The pots were formed and fired in Germany at a firing temperature of 850 °C, a heating rate of 2.3 K/min and a retention time of 30 min.

Leaching effects were again analyzed applying a series of 23 x 1 L tap water on each filter pot. An analysis of filtrates 1, 5 and 7 for arsenic and manganese and a broad



analysis for filtrate 2 of each filter pot were done at the *CIP Chemisches Institut Pforzheim GmbH*.

**Effect of fractioned filtration.** Subsequently, natural water was applied to the filter pots in order to determine further effects of the filter on the filtered water. Therefore, water with high organic and bacterial load of a small brook (Alte Bach, Stutensee, Germany, samples taken on October 7<sup>th</sup> 2014) and a big river (River Rhine at kilometer 362.07, Germany, samples taken on October 14<sup>th</sup> 2014) were applied to the filter pots. The experiment was conducted in duplicate with a separate bucket of natural water for each set of experiment. To detect the effect of a fractioned filter, about 6 L of the water was first applied to the filter with the highest porosity meaning the filter with 40 % rice husks. About 2 L of the filtrate were kept for further analysis, while the rest was applied to the pot with 20 % rice husks. Again 2 L were kept for analysis and the remaining 2 L were applied to the pot without rice husks and with the smallest porosity. Micro as well as a molecular biological analysis was done to detect effects of the filter pots on the bacterial population of the natural water samples. Therefore, duplicates of 100 mL samples were filtered through mixed cellulose ester and poly-carbonate membranes (both 0.2  $\mu\text{m}$ ,  $\varnothing$  47 mm, Whatman<sup>TM</sup> GmbH, Lawrence, USA) and analyzed for pseudomonads, coliform bacteria and enterococci using species specific agar.

HPCs were determined according to chapter 2.1.4. Moreover, 100 mL of each sample was filtered through a poly carbonate membrane which was directly used for DNA extraction. A population analysis with PCR and DGGE was performed according to chapter 2.1.5. Physico-chemical performance of raw water and filtered water was performed at the *CIP Chemisches Institut Pforzheim GmbH* (chapter 2.1.3). Turbidity was determined in triplicate using a portable turbidity meter Turb® 355IR/T (WTW GmbH, Weilheim, Germany).

The flow rate was noted throughout the experiments.

Following the experiments with natural river water, microbiological performance of the filters was further examined applying tap water spiked with *E. coli*, *P. aeruginosa* and *Ent. faecium*. Therefore, cultures were cultivated as previously described and mixed with 2 L of tap water according to chapter 2.1.4. A 1 L aliquot of this raw water was applied to the filter containing 40 % rice husks. While 200 mL of the filtrate were kept for analysis, 800 mL of the filtrate were applied to the filter pot containing 20 % rice husks. Again

200 mL of the filtrate were kept for analysis and 600 mL were applied to the pot without rice husks. The filtrate was kept for analysis. Raw water and filtrate were examined as described in chapter 2.1.4 to detect the log reduction.

Moreover, the effectiveness in phage removal was detected. Therefore, 3 L of tap water were spiked with a  $\phi$ X174 phage suspension and all but 600 mL of this raw water was applied to the filter containing 40 % rice husks. The filtrate was applied to the filter pot containing 20 % rice husks and the filtrate of this filter was again applied to the pot containing no rice husks. Of each filtrate and the raw water 600 mL were kept for analysis. In each case, 500 mL of these samples were concentrated for phage detection as described in chapter 2.1.4.

The experiments were repeated with filters made of the same concentration of sawdust.

**Effect of silver coating.** In order to figure out if the application of silver significantly improves the antimicrobial effectiveness, a filter pot containing 20 % sawdust was submerged in a 1.26 g/L silver nitrate solution for 1 min. First, the leaching effect of the silver was detected. Therefore, a series of 22 x 1 L tap water was applied to the coated filter as well as an uncoated filter of the same composition. Physico-chemical analysis of the filtrates was conducted as previously described. After this physico-chemical analysis, microbial performance was analyzed.

## **2.4 Summary**

As a basis for developing an optimized filter, preliminary studies of an existing filter were conducted in the field and lab to identify drawbacks and optimization potential for the use in Gunungkidul. In detail, morphology, mineralogical and chemical composition as well as physico-chemical and microbiological performance were analyzed in the laboratory. In the field study, performance, acceptance by the population and ease of operation and maintenance were observed.

To develop an appropriate firing procedure and composition, local firing habits and temperature profiles as well as available raw material were analyzed. Apart from the traditionally used clay mixture, several clay sources, lava sand components and combustible material used for forming pores were examined. The analysis program comprised composition of raw material, influence of firing temperature on morphology, influence of combustible material on pore size distribution and porosity as well as physico-chemical and microbial performance. Fractioned filtration and application of a silver coating were examined for influence on bacteria and phage removal.

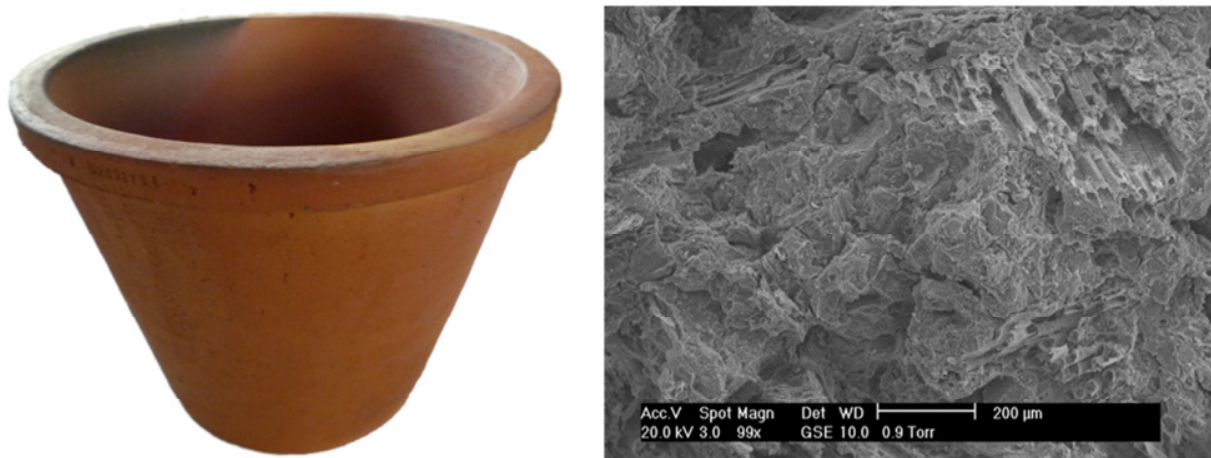
### 3 Results

In this dissertation, an appropriate composition and firing procedure was developed for the local production of ceramic filters for POU drinking water treatment in Gunungkidul. As a start, an existing *PFP* filter (with silver treatment) produced by *Pelita* in another region in Indonesia was analyzed in order to identify issues that needed to be addressed in developing a filter optimized for the use in Gunungkidul.

#### 3.1 Analysis of the existing filter produced by *Pelita* as a basis for optimization

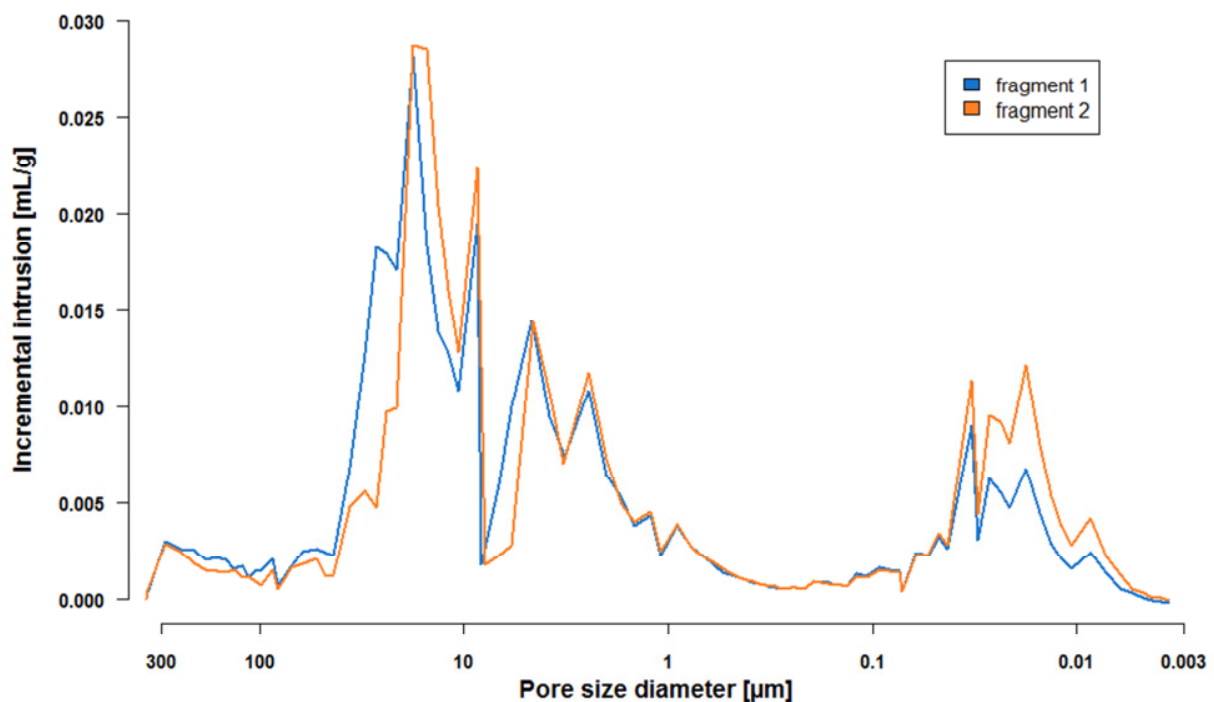
**Morphology.** The filter produced by *Pelita* is shown in Figure 11. It had a reddish color, was frustum shaped and had a capacity of about 7.8 L. The filter wall had a thickness of 1.6 cm at the sides and 3.3 cm at the bottom. The inner height of the filter was about 18.5 cm and the angle between the sides of the filter and the vertical axis was  $43.2^\circ$ . The bottom had a diameter of 15.7 cm.

ESEM pictures showed that the filter surface (figures in appendix, Figure A 3) was relatively smooth. The broken edge of the filter (Figure 11) showed characteristic ceramic pores and crystal structures, as well as tiny, tube-like structures produced when pore-forming agents like sawdust or rice husks burned out completely during the firing process.



**Figure 11:** Shape of the Pelita filter (left) and ESEM picture of breaking edge of the filter (right).

Mercury porosimetry was used to determine pore size distribution. Two different fragments of a filter were analyzed. Both fragments had a total porosity of about 49 %. The fragments showed a substantial group of peaks between 1 to 40  $\mu\text{m}$  with a smaller group from 0.006 to 0.04  $\mu\text{m}$  (Figure 12).



**Figure 12:** Pore size distribution.

**Mineralogical and chemical composition.** A qualitative X-ray analysis of the filter showed that existing phases were quartz ( $\text{SiO}_2$ , trigonal), hematite ( $\text{Fe}_2\text{O}_3$ , trigonal), tridymite ( $\text{SiO}_2$ , rhombic) and mullite ( $\text{Al}_6\text{Si}_2\text{O}_{13}$ , rhombic), which corresponded to ED XRF results (Table 5). Moreover, ED XRF showed that some metals were present in relatively high concentrations and could, potentially, have a negative effect on human health if leached out in high concentrations. They were, particularly, As, Ba, Cd, Cr, Mn, Pb and Zn.

**Table 5:** Elemental composition of the Pelita filter with main components and selected trace elements.

\*filter fragment was not coated with silver

Main components	concentration	Trace elements	concentration
Al <sub>2</sub> O <sub>3</sub>	21 %	As	15 ppm
SiO <sub>2</sub>	64 %	Ba	250 ppm
K <sub>2</sub> O	1.31 %	Cr	82 ppm
CaO	0.78 %	Cu	29 ppm
TiO <sub>2</sub>	1.01 %	Ni	14 ppm
MnO	0.18 %	Pb	30 ppm
Fe <sub>2</sub> O <sub>3</sub>	10.9 %	Zn	116 ppm

**Physico-chemical performance.** To analyze the filter's chemical performance, several parameters were measured in raw and filtered water. Initially, the effect of the filter on laboratory tap water was analyzed. Results are shown in Table 6. Tap water properties corresponded to the German drinking water regulation (DVGW, 2001). The pH of the filtered water was slightly higher (pH 8) than the raw water (pH 7). No significant changes were noted in electrical conductivity (EC), hardness or nitrite concentration. For nitrate, there was a peak of 12 mg/L in the first filtrate F1, but subsequent elutions showed no concentrations that were significantly higher than those in the raw water. Arsenic, manganese and silver were all leached from the filter in significant concentrations. Arsenic was present at about 0.09 mg/L initially, although concentrations decreased in subsequent batches of filtrate. In F7, the concentration was down to 0.018 mg/L. Manganese was leached from the filter with a peak of 0.2 mg/L in the first filtrate, F1, and rapidly decreasing concentrations after that. Silver concentrations in the filtrate were about 0.01 to 0.03 mg/L, and were up to 26 times higher than those in the raw water. Concentrations of barium in the filtered water were about 0.03 mg/L and thus lower than in raw water (0.14 mg/L). Concentrations of chromium and cadmium in filtered water did not differ significantly from raw water. Lead was leached from the ceramic in low concentrations (0.003 mg/L) whereas zinc was reduced from 0.03 mg/L in tap water to 0.001 mg/L in the filtrate.

**Table 6:** Measured parameters for tap water (TW) and selected filtrates (F). WHO gives the WHO guideline values. \*Initial flow rate (first liter). \*\*Not of health concern at levels found in drinking water. \*\*\*Available data inadequate to permit derivation of health-based guideline value. \*\*\*\*health-based value

parameter	WHO	TW	F1	F2	F3	F4	F5	F6	F7
Volume [L]	-	-	3	3	5.5	5.5	5.5	5.5	5.5
Flow rate* [L/h]	-	-	-	0.73	1.82	1.82	1.20		
pH [-]	**	6,98	7.99	8.03	7.93	8.03	8.06	8.03	7.96
T [°C]	-	18.1	17.4	20.2	19.8	20.9	20.3	20.2	20.2
EC [µS/cm]	-	560	511	525	528	529	-	-	-
Turbidity [NTU]	1	0.15	0.18	0.31	0.17	0.17	-	-	-
Hardness [mg/L CaCO <sub>3</sub> ]	**	2.93	2.68	2.81	2.90	2.93	-	-	-
Nitrate NO <sub>3</sub> <sup>-</sup> [mg/L]	50	2.9	12.14	3.08	3.17	2.76	3.01	2.64	2.56
Nitrite NO <sub>2</sub> <sup>-</sup> [mg/L]	3	<0.01	-	0.01	-	0.01	-	-	-
Ag [mg/L]	***	0.001	0.026	0.015	0.011	0.016	0.022	0.020	0.023
As [mg/L]	0.01	-	0.08	0.089	-	0.049	0.067	-	0.018
Ba [mg/L]	0.7	0.139	0.038	0.029	0.032	0.033	0.035	0.048	0.060
Cr [mg/L]	0.05	0.006	0.005	0.004	0.004	0.005	0.005	0.005	0,005
Cd [mg/L]	0.003	<0.001	<0.001	0.001	<0.001	<0.001	<0.001	-	-
Mn [mg/L]	0.4****	0.002	0.222	0.010	0.017	0.001	0.005	0.004	0.004
Pb [mg/L]	0.01	-	-	0.003	-	<0.002	-	-	-
Zn [mg/L]	**	0.027	0.001	0.001	0.002	0.001	0.001	0.001	0.001
TOC [mg/L]	-	0.56	-	1.5	-	2.0	-	0.46	-

The second set of experiments was carried out with natural river water and the results resembled those from tap water. Furthermore, there was a significant reduction in turbidity from 2.3 NTU (raw) to 0.1 NTU (filtrate) – see the first part of Table 7 (columns headed RW and RF).

Field study experiments were conducted with Indonesian raw water (IW) using a brand new and a two-year-in-use *Pelita* filter. The second part of Table 7 – columns IW, IF-old and IF-new – shows the results of these experiments in Indonesia. While 7.5 L were applied to the filters, the old filter, in daily use for about two years, had a flow rate of about 0.1 L/h. The new filter's initial flow rate was about 1.2 L/h. Complying with the laboratory experiments, turbidity, TOC, arsenic, manganese, nitrate and nitrite were washed from the new filter in the first filtrate. It is remarkable that the old filter reduced the EC of the filtered water from 504 to 270  $\mu\text{S}/\text{cm}$ , while turbidity was reduced to 0.5 NTU and TOC to 0.2 mg/L (complete data in appendix, Table A 1).

**Table 7:** Analyte concentrations for river water RW in Germany and field study in Indonesia.

(RF = river water filtrate, IW = Indonesian raw water, IF old = filtrate from old filter, IF new = first filtrate from new filter. \*Initial flow rate (first liter))

Parameter	Germany		Indonesia		
	RW	RF	IW	IF old	IF new
Volume [L]	-	6	-	7.5	7.5
Flow rate* [L/h]	-	0.75	-	0.1	1.2
pH [-]	7.66	7.92	7.4	7.95	7.54
EC [ $\mu\text{S}/\text{cm}$ ]	470	457	504	270	519
Turbidity [NTU]	2.30	0.14	1.2	0.52	8.79
Hardness [mg/L $\text{CaCO}_3$ ]	1.57	1.62	2.83	1.55	2.1
Nitrate $\text{NO}_3^-$ [mg/L]	7.71	7.80	8.3	9.7	21
Nitrite $\text{NO}_2^-$ [mg/L]	0.08	0.09	<0.01	<0.01	25.5
As [mg/L]	<0.001	0.045	<0.001	0.001	0.08
Mn [mg/L]	0.003	0.001	<0.01	<0.01	0.27
TOC [mg/L]	1.99	2.15	0.72	0.22	23

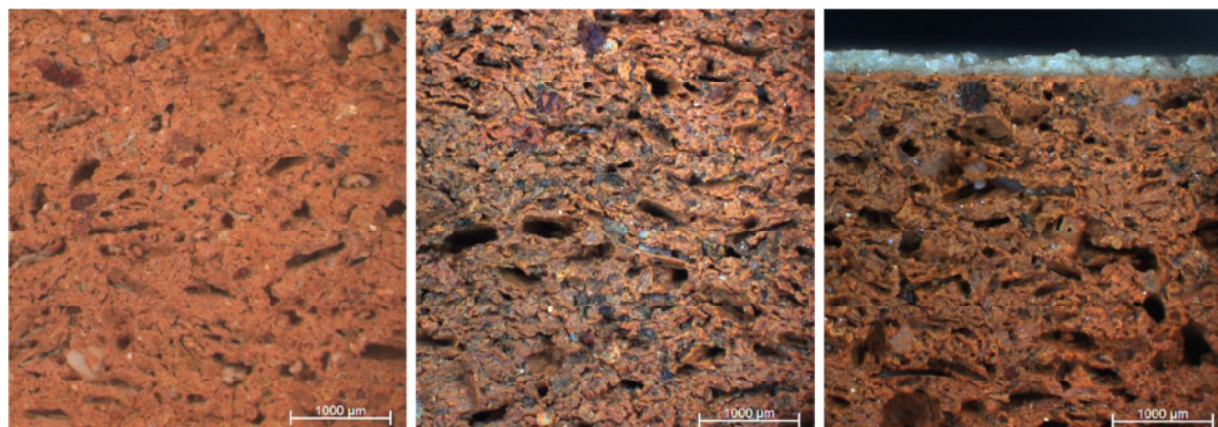


**Flow rate and hydraulic conductivity.** During the tap water experiments, the initial flow rate dropped from 1.8 L/h (F3) to 1.2 L/h (F5) while 5.5 L were applied to the filter for each measurement. The flow rate for natural river water was about 0.75 L/h when 6 L were applied, but it was found that the flow rate increased again when less turbid water was applied.

After 100 L of tap water had been filtered, the initial flow rate was about 1.4 L/h but it dropped to 1 L/h after 200 L were filtered. Following the equations of Schweitzer *et al.* describing the flow rates for the bottoms and sides of ceramic filters (Schweitzer *et al.*, 2013), and based on Darcy's equation, the hydraulic conductivities ( $k$ ) of the filters in this study were between 1.5 and  $4.4 \cdot 10^{-7}$  m/s. After 300 L were filtered, a thorough flow rate analysis depending of water height in the filter was conducted and a median flow rate of 0.6 L/h with a hydraulic conductivity of  $1.4 \cdot 10^{-7}$  m/s was calculated.

Field observations showed that after 1 year (330 days) of daily use in Gunungkidul and an estimated volume of about 8,320 L filtered water, the initial flow rate decreased to 0.8 L/h.

**Fouling.** After about two years (745 days) of daily use (total about 13,300 L) in Gunungkidul, the initial flow rate was down to about 0.2 L/h indicating that fouling had occurred. By eye, no biofilm was visible at the filter surface. Further analysis was performed in Germany, but due to transportation regulations, filters could only be shipped in a dehydrated condition. Incident light microscopy showed a layer of salt crystals at the inner filter surface and additional crystals within the pores. In Figure 13, an almost unused filter and the used filter are shown. The filters had slightly different colors due to different quantities of feldspars such as andesines.



**Figure 13:** Incident light microscopy of almost new filter (left) and used filter (middle & right).

XRF spectroscopy showed no significant differences between unused and used filter, as variations between filters were too high to allow a direct comparison. Loss on ignition of the used filter was about 1.3 % indicating low amount of organic material in the used filter.

**Microbiological performance.** Bacteria reduction experiments showed that the filter was able to remove about 5 log of *E. coli*, 3.4 to 4.5 log of *Ent. faecium* and 3.4 to 5 log of *P. aeruginosa*. Bacteria counts in the raw water and filtered water are given in Table 8.

**Table 8:** Bacteria count [CFU/mL] and <sup>7</sup>log removal for *E. coli*, *Ent. faecium* and *P. aeruginosa* (RAW = raw water, F = filtrate)

Bacteria count [CFU/mL]	First experiment			Repetition		
	<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeruginosa</i>	<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeruginosa</i>
RAW	$2.4 \cdot 10^6$	$3.4 \cdot 10^4$	$8.7 \cdot 10^5$	$9.9 \cdot 10^6$	$2 \cdot 10^6$	$1.8 \cdot 10^7$
F	$2 \cdot 10^1$	0	$3.5 \cdot 10^2$	$6 \cdot 10^1$	$8.8 \cdot 10^2$	$2 \cdot 10^2$
<b>Log removal</b>	<b>5.1</b>	<b>&gt;4.3</b>	<b>3.4</b>	<b>5.2</b>	<b>3.4</b>	<b>5.0</b>

Quantitative PCR results showed a log removal of 1.7 to 2.5 for *E. coli*, 2.8 to 3.4 for *Ent. faecium* and 1.5 to 2.0 for *P. aeruginosa*.

<sup>7</sup> Log removal =  $\log_{10}$  (bacteria count in RAW / bacteria count in F)

Phage reduction experiments showed that the log removal was about 0.3 to 0.6 log for  $\phi$ X174 and 0.4 to 0.8 log for MS2. Turbidity in the tap water experiments was lower than 1 NTU. In experiments with higher turbidity (8.5 NTU), the log reduction for  $\phi$ X174 was increased to 0.9 (see Table 9).

**Table 9:** Plaque forming units [PFU/mL] and log removal (RAW = raw water, F = filtrate)

Plaque count [PFU/mL]	First experiment		Repetition		Turbidity 8.5 NTU
	$\phi$ X174	MS2	$\phi$ X174	MS2	$\phi$ X174
RAW	$1.4 \cdot 10^3$	$7.9 \cdot 10^3$	$2.6 \cdot 10^2$	$7.9 \cdot 10^3$	$5.1 \cdot 10^3$
F	$3.2 \cdot 10^2$	$1.4 \cdot 10^3$	$1.2 \cdot 10^2$	$3 \cdot 10^3$	$6.6 \cdot 10^2$
<b>Log removal</b>	<b>0.6</b>	<b>0.8</b>	<b>0.3</b>	<b>0.4</b>	<b>0.9</b>

Original paper on the *Pelita Indonesia* filter experiments published in: Matthies K., Bitter H., Deobald N., Heinle M., Diedel R., Obst U., Brenner-Weiss G. (2015) *Morphology, composition and performance of a ceramic filter for household water treatment in Indonesia*. Water Practice and Technology (doi: 10.2166/wpt.2015.044).

### 3.2 Evaluation of handling of ceramic filters

To evaluate if ceramic filters are appropriate for use in Gunungkidul, field studies were done in which three local people were given ceramic filters produced by *Pelita*. They were introduced to the filter and got training in handling and maintenance. Within the duration of the project, the users were frequently questioned and observed when handling the filter.

In general, the ceramic filter was very well accepted. Locals found the operation of the filter easy and according to them, sufficient water was treated when they filled the filter in the morning before going to work in the field and then again when they came back. They said they would be happy to use ceramics as their ancestors used to store their water in pottery to keep the water cool. Moreover, users said that tea tasted better after filtering the water and tea color was brighter from filtered water compared to tea from raw water. They also reported that less lime deposited in the cooking pots when the water was filtered before boiling (see appendix, Figure A 4).

During the field studies, it became obvious that the most critical part for use of the filter is the cleaning. Though users were told to clean the filter regularly (at least once a month), filters were cleaned only rarely.

If the filter is cleaned at all, the users have to take the filter out of the receptacle. Therefore, users touch the outer “clean” side of the filter and might also put the filter on a surface that has not been cleaned before. Thus, a contamination of the “clean” side of the filter is likely. An impression of how the filter is cleaned under local conditions can be seen in the appendix (Figure A 5).

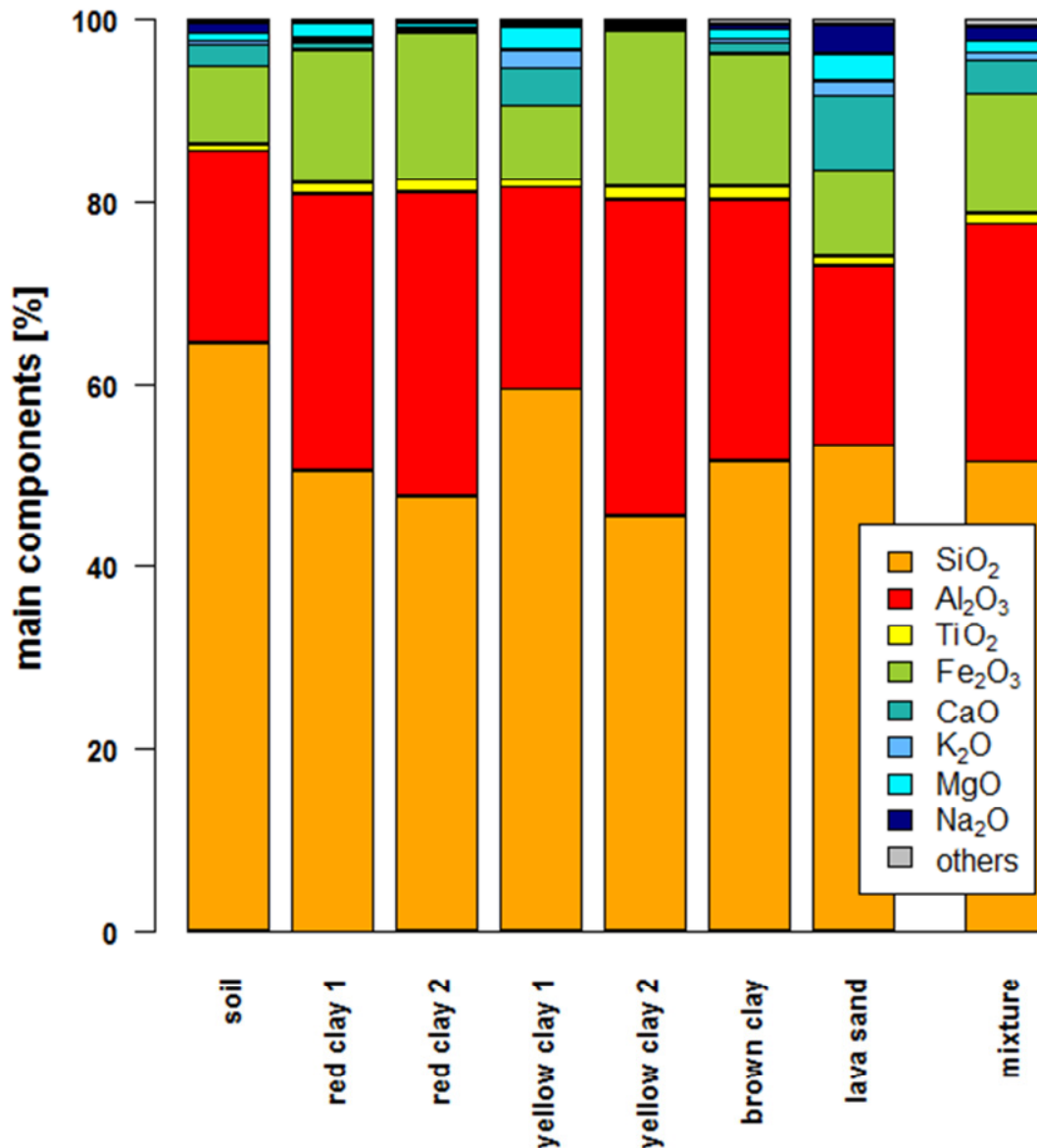
### **3.3 Development of an optimized filter to be locally produced**

To develop a composition and firing procedure for the local production of appropriate ceramic filters in Gunungkidul, an analysis of locally available material, local firing habits as well as physico-chemical and microbiological performance needed to be conducted. The results of these analyses are given in the next paragraphs.

#### **3.3.1 Analysis of local raw material**

**Chemical composition.** To determine the composition of the local clays, the elementary oxides present in the traditional clay mixture as well as raw materials used in this mixture were analyzed.

As shown in Figure 14, the three main components of all samples were silicon dioxide  $\text{SiO}_2$  (silica) with about 45.5 % to 64.5 %, aluminum oxide  $\text{Al}_2\text{O}_3$  (alumina) with 19.8 to 34.7 % and iron (III) oxide  $\text{Fe}_2\text{O}_3$  with 8.5 to 17.1 %. Lava sand, soil, yellow clay 1 and the clay mixture showed a remarkable concentration of calcium oxide  $\text{CaO}$  (calcia) with 2.4 % in soil to 8.2 % in lava sand. Soil, lava sand and the clay mixture additionally contained remarkable concentrations of sodium oxide  $\text{Na}_2\text{O}$  with 1.1 to 3.2 %. Titanium dioxide  $\text{TiO}_2$  (titania) was present in every sample with 0.8 to 1.5 %. Highest concentrations of magnesium oxide  $\text{MgO}$  (magnesia) was present in red clay 1, yellow clay 1 and lava sand with concentrations of 1.75 to 2.83 % (complete data in appendix, Table A 2).



**Figure 14:** Main components of the local raw materials used in the traditional mixture.

It is remarkable that the samples red clay 2 and yellow clay 2 were very similar regarding their composition, while red clay 1 and brown clay showed a similar composition. In contrast, yellow clay 1 and 2 were quite different in their composition.

The clay mixture produced from changing clay sources is not attained following a specific composition but changes according to raw materials used. The mixture is adjusted according to the feeling of the potters. On one sampling day the mixture was attained by mixing some red clay with soil and lava sand. During the second observation, the mixture composed of red clay 1, yellow clay 1, soil and lava sand

(August 2013) and on the third occasion red clay 2, brown clay and lava sand were mixed. A few days later the mixture consisted of red clay 2, yellow clay 2 and lava sand.

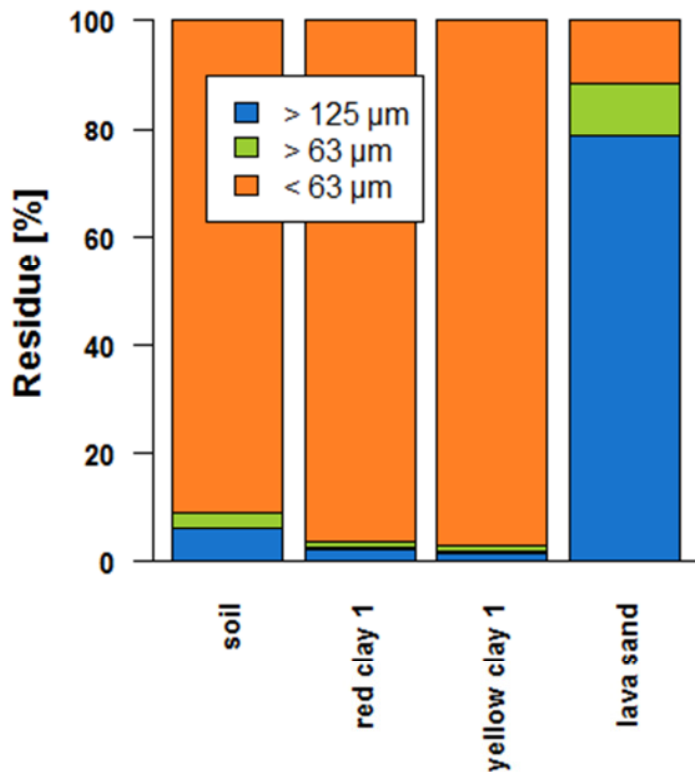
As arsenic and manganese leaching were detected in previous ceramic filters, ICP-OES of an aqua regia digestion of the raw material was conducted. Results (complete data in appendix, Table A 3) showed that arsenic could not be detected in high concentration (<3 mg/kg (sawdust) to 8 mg/kg (yellow clay)). However, manganese was present in all samples with concentrations of 30 (sawdust) to 1180 mg/kg (soil).

**Mineralogical composition.** Mineral phases detected in the samples included hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) and maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ), calcite ( $\text{Ca}[\text{CO}_3]$ ) and a huge group of quartz and silicates. Calcite was detected in yellow clay 1. Hematite was detected in the red clay samples, yellow clay 2 and brown clay, while maghemite was found in red clay 2 and yellow clay 2. Quartz ( $\text{SiO}_2$ ) was found in the red clay samples, yellow clay samples and brown clay, while cristobalite ( $\text{SiO}_2$ ) and kaolinite ( $\text{Al}_2[(\text{OH})_4\text{Si}_2\text{O}_5]$ ) were present in red clay 2, yellow clay 2 and brown clay. Moreover, in red clay 1, nacrite (kaolinite) and other not further specified mineral phases were detected. Yellow clay 1 composed of illite (hydro muscovite) ( $(\text{K}, \text{H}_3\text{O}) \text{Al}_2[(\text{OH})_2/\text{AlSi}_3\text{O}_{10}]$ ) and vermiculite.

Soil composed of anorthite (plagioclase, feldspar) ( $\text{Ca}[\text{Al}_2\text{Si}_2\text{O}_8]$ ) and nontronite (smectite) which often show a high proportion of iron and other mineral phases.

All mineral phases detected in lava sand belonged to the quartz and silicate group. The sand composed of andesine (anorthite, plagioclase, feldspar), cordierite ( $(\text{Mg}, \text{Fe})_2\text{Al}_3[\text{AlSi}_5\text{O}_{13}]$ ) and diopside ( $\text{CaMg}[\text{Si}_2\text{O}_6]$ ).

**Grain size distribution.** In order to get an impression on the grain size distribution, a wet sieving was done. As shown in Figure 15, soil as well as both red and yellow clay had a huge fraction below 63  $\mu\text{m}$  grain size which was about 91 % in soil and about 97 % in red and yellow clay. The fraction between 63  $\mu\text{m}$  and 125  $\mu\text{m}$  was relatively small for all three samples with 1.1 to 3.1 % whereas the fraction above 125  $\mu\text{m}$  was about 1.6 to 6 %. In contrast, lava sand had an 11.6 % fraction below 63  $\mu\text{m}$ , 9.7 % between 63 and 125  $\mu\text{m}$  and 78.7 % above 125  $\mu\text{m}$ .



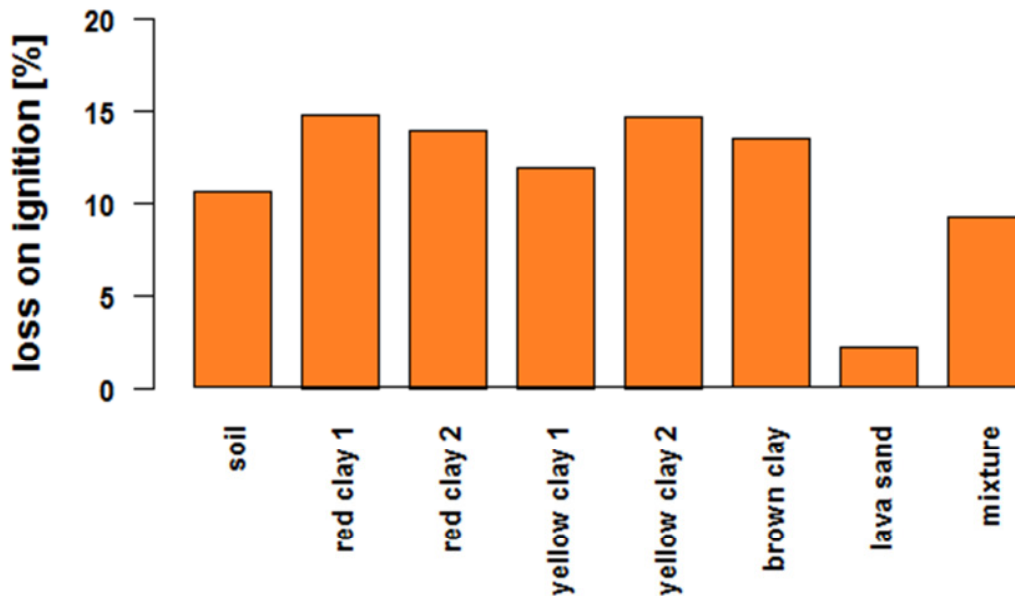
**Figure 15:** Grain size distribution of samples.

As the fractions below 63  $\mu\text{m}$  were quite big for soil, red and yellow clay, light-scattering technique was used to further analyze the distribution within this fraction. In Table 10, the results are shown. 99.9 % of the grains had a size of less than 69.3  $\mu\text{m}$  for soil, less than 42.7  $\mu\text{m}$  for red clay 1 and 58.8  $\mu\text{m}$  for yellow clay 1. For lava sand, 99.9 % had a grain size of 184  $\mu\text{m}$ . 10 % of the grains still had a diameter of less than 0.8  $\mu\text{m}$  for red clay 1, less than 1.1 to 1.3  $\mu\text{m}$  for yellow clay 1 and soil and less than 1.9  $\mu\text{m}$  for lava sand.

**Table 10:** Grain size distribution within the fraction below 63  $\mu\text{m}$   
 $D_{10}$  is the diameter at which 10 % of the grains have a smaller grain size.

Sample	$D_{10}$ [ $\mu\text{m}$ ]	$D_{50}$ [ $\mu\text{m}$ ]	$D_{90}$ [ $\mu\text{m}$ ]	$D_{99.9}$ [ $\mu\text{m}$ ]
soil	1.27	4.66	28.0	69.3
red clay 1	0.843	2.75	12.5	42.7
yellow clay 1	1.11	3.17	13.0	58.8
lava sand	1.94	25.7	84.8	184

**Loss on ignition** of red, yellow and brown clays varied between 12 % for yellow clay 1 and 14.8 % for red clay 1. Soil showed a 10.6 % loss on ignition, while lava sand had a significantly lower value of 2.3 % corresponding to the low amount of clay minerals and bound water of crystallization. In the mixture, this resulted in a loss on ignition of 9.3 % (Figure 16).



**Figure 16:** Loss on ignition [% w/w] of the local raw materials used in the traditional clay mixture.

### 3.3.2 Analysis of local firing process

The local pottery village Kasongan is one of the most important pottery centers in Indonesia (Kementerian Perdagangan, 2014).

There are very few potteries in Kasongan using gas-fired car kilns with a temperature of about 1050 °C. In these kilns traditional figures for tourists are produced. The kilns could also be used at lower firing temperatures. Using these kilns for production of ceramic filters would enable a good reproducibility. However, a study comparing costs for production of ceramic filters in these gas-fired kilns and traditional wood-fired kilns would be necessary.

Most of the potteries use traditional wood-fired kilns such as the one shown in the appendix (Figure A 6). It is made from bricks that form a square with an opening at the front. There is no fixed top or even a chimney. Wood and thus fire is introduced in chambers below the floor level and heat is drawn up. It has to be mentioned, that the wood quality differs depending on availability and price. Normally, only cheap waste

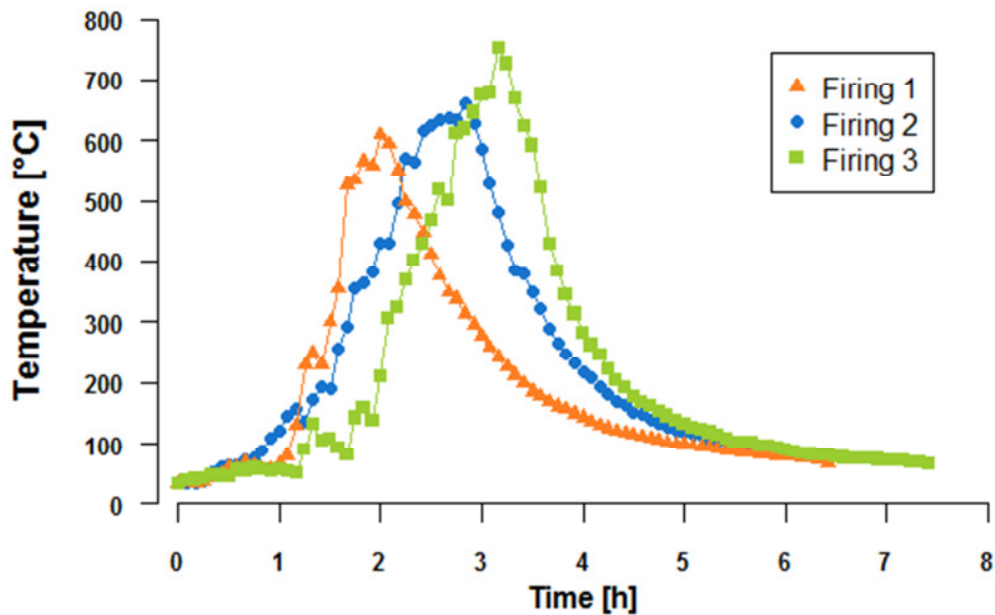


wood from furniture craftsmen is used and all waste from the potter's household including plastics is burned alongside the wood which additionally influences the heating performance.

Prior to the firing process, the kiln is covered using metal rods as supporting grid, roof tiles, bricks, metal sheets and straw as shown in the appendix (Figure A 7).

According to local potters, the firing temperature is not measured during firing but they estimated that it is about 700 to 800 °C (personal communication). Thus, in these traditional potteries, earthenware pottery is produced such as flower pots or traditional stoves. For the firing process, potters are not following a certain protocol. They are adding wood to the fire according to their experience. Typically, the ware is first dried for about two to four days depending on the season. Then it is stacked in the kiln and slowly fired until the potter decides to start the "big" fire which lasts for about two hours. When this fire is finished, no more wood is added and the kiln is allowed to cool down before it is opened again. All in all, the firing process takes about 10 to 12 hours (personal communication).

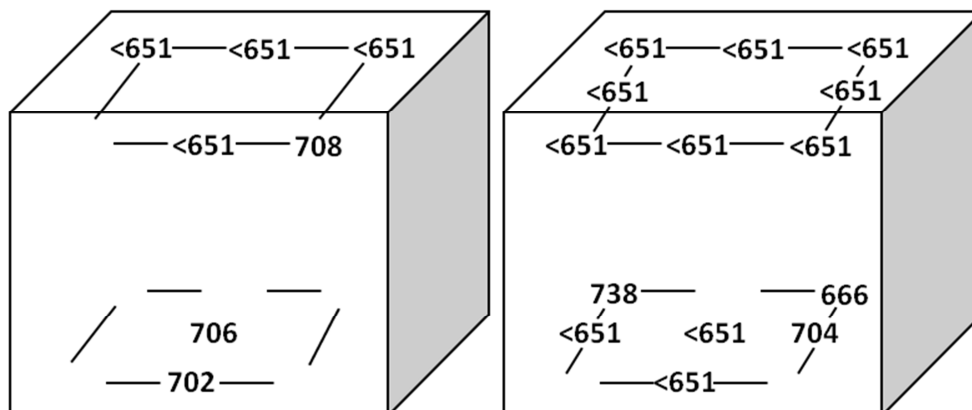
In Figure 17, three temperature profiles of separate firing events are shown. During the first firing the temperature rose from 79.8 °C to 526.3 °C in 35 min before further rising to a maximum of 608.4 °C in 20 min. The second firing event showed a temperature increase from 137.1 °C to 616.5 °C in 70 min and a further increase to the maximum of 659.6 °C in 25 min. In the third firing the temperature rose from 79 °C to 751.4 °C in 90 min.



**Figure 17:** Temperature profiles of three separate traditional firing processes.

Considering only the relatively linear heating phase, heating rates varied from 6.9 K/min in the second firing to 12.8 K/min in the first firing.

In addition, the distribution of the maximum temperature throughout the firing kiln was determined. Results are shown in Figure 18. The left side of the figure corresponds to the first measurement in Figure 17, while the right side corresponds to the second measurement. In general, the temperature at the bottom of the kiln is higher compared to the top. Variations between top and bottom as well as between firing events were more than 50 °C.

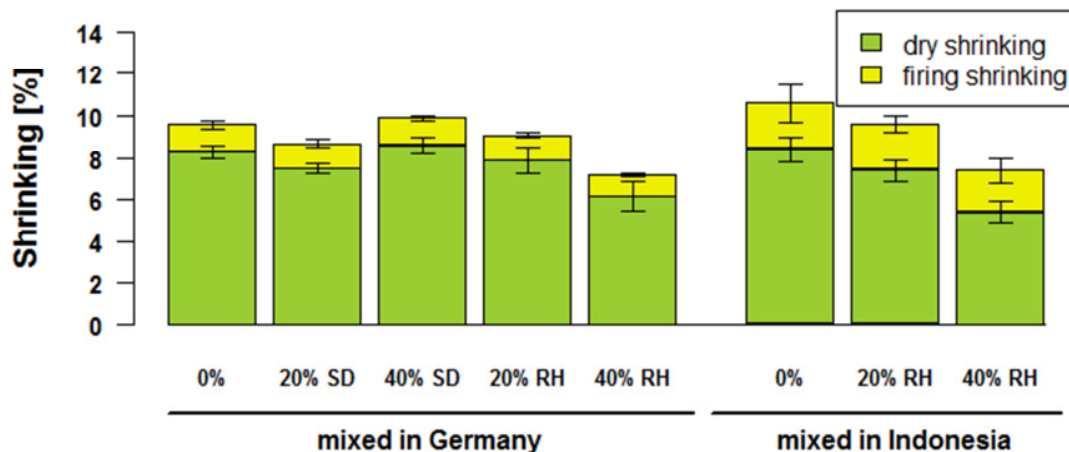


**Figure 18:** Distribution of maximum temperature throughout the kiln during two traditional firing events. 650 °C was the lower detection limit for the temperature control rings used in the experiments.

### 3.3.3 Development of an appropriate composition

For developing an appropriate composition using locally available material, the traditional clay mixture was mixed with sawdust and rice husks in concentrations of 0, 20 and 40 % of combustible material. These mixtures were fired and analyzed regarding shrinking behavior, bulk density, water absorption, porosity and pore size distribution. Moreover, the results were compared to mixtures that were attained directly at the potteries in Indonesia, containing 0, 20 and 40 % of rice husks.

**Shrinking.** As shown in Figure 19, dry shrinking decreased with increasing concentration of combustible material from 8.3 % for the mixture not containing combustible material to 6.2 % for the sample containing 40 % rice husks. However, the sample containing 40 % sawdust was an exception as its' shrinking during drying was even slightly higher (8.6 %) than for the sample not containing any combustible material (8.3 %). The shrinking during firing was relative constant with 1 to 1.3 % for the samples mixed in Germany. The samples mixed in Indonesia reacted similar with a slightly higher firing shrinking of 2 to 2.2 %.

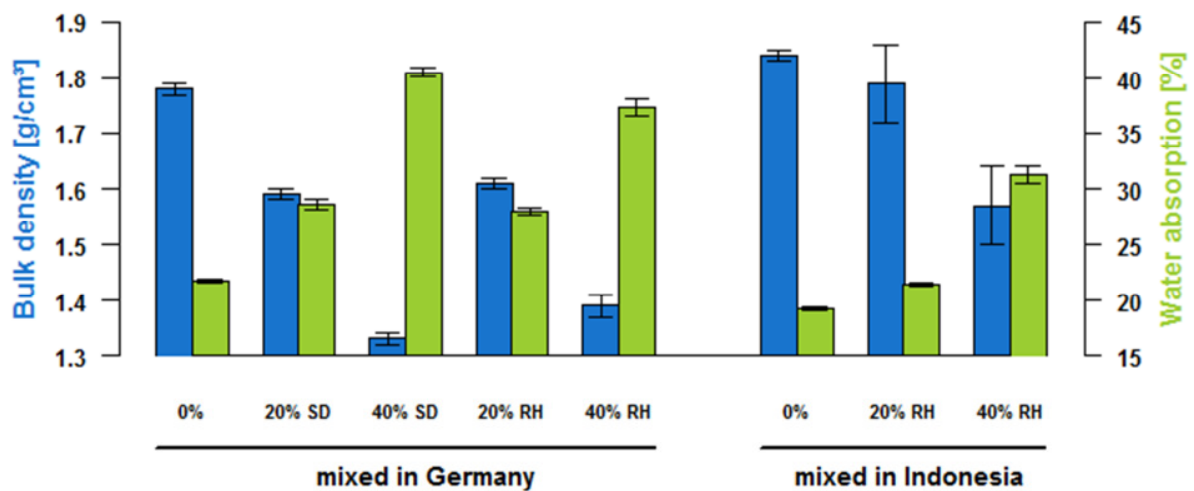


**Figure 19:** Dry and firing shrinking of clay mixtures composing of 0, 20 and 40 % sawdust (SD) or rice husks (RH).

**Bulk density and water absorption.** With increasing concentration of combustible material, the bulk density (blue bars in Figure 20) of the samples decreased from 1.8 for the sample without combustible material to 1.3 g/cm<sup>3</sup> for the sample containing 40 % sawdust. The decrease was slightly higher for sawdust compared to rice husks as

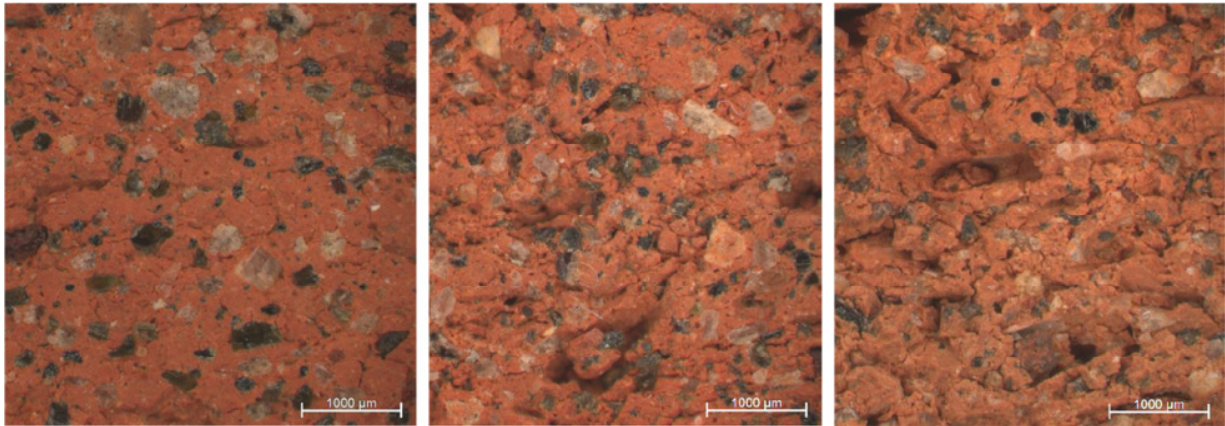
combustible material. The samples that were mixed in Indonesia showed the same trend, though the decrease was not as high.

Accordingly, the water absorption (green bars in Figure 20) increased with increasing content of sawdust or rice husks from 22 % for the pure clay mixture to 40 % for the mixture containing 40 % sawdust. Again, the increase was slightly higher for sawdust compared to rice husks. Samples mixed in Indonesia reacted similar with a lower increase compared to samples mixed in Germany.



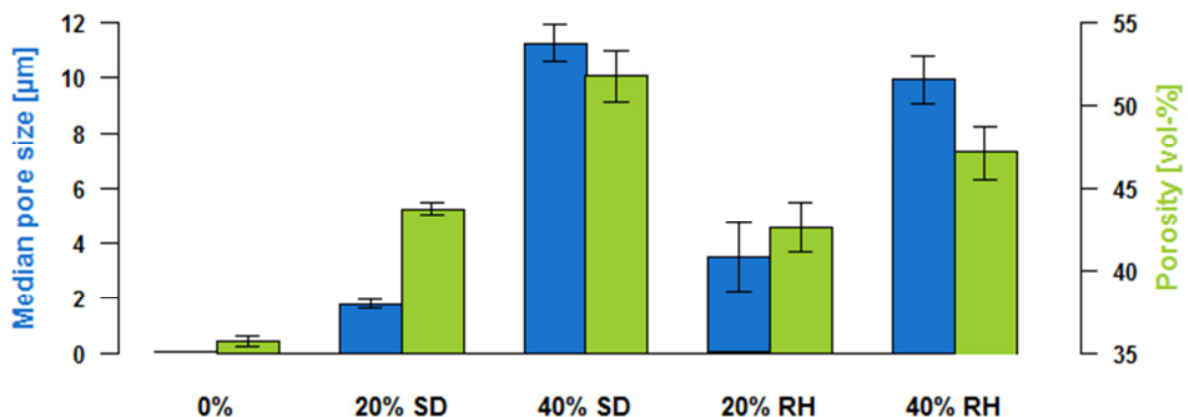
**Figure 20:** Bulk density and water absorption of clay mixtures composing of 0, 20 and 40 % sawdust (SD) or rice husks (RH).

**Porosity.** Figure 21 shows the cut surfaces of ceramics containing 0 %, 20 % and 40 % rice husks. Pores formed by burning of the combustible material are not homogeneously distributed and vary in size. While the cut surface of the sample containing 0 % rice husks is quite smooth, the ceramic made of 40 % rice husks shows a very rough cut surface. Black and grey particles visible in the ceramic are minerals such as feldspars that have not been transformed during firing.



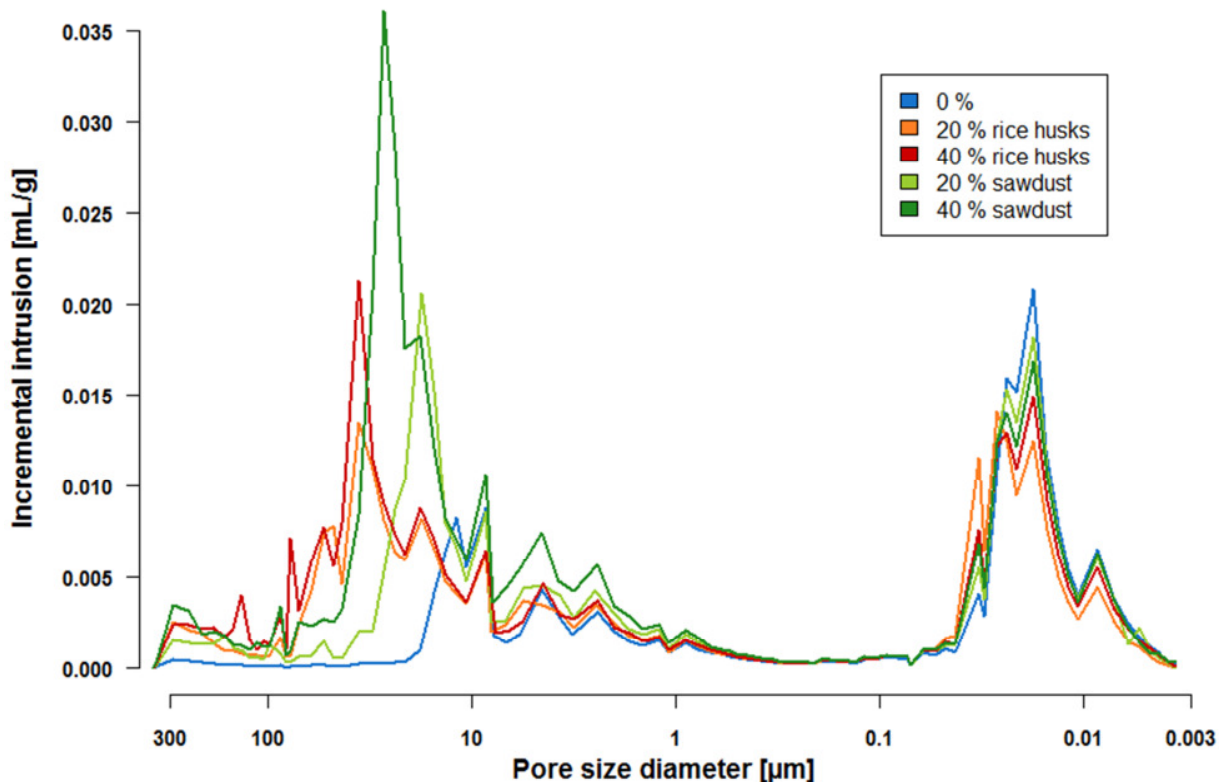
**Figure 21:** Incident light microscopy of ceramics containing 0 % combustibles (left), 20 % rice husks (middle) and 40 % rice husks (right).

In Figure 22, median pore size (blue bars) and porosity (green bars) of the samples containing different levels of combustible material are shown. Both, median pore size and porosity increased with increasing level of sawdust or rice husks from 0.03  $\mu\text{m}$  pore size and 36 % porosity in the sample without pore-forming agents to 11.24  $\mu\text{m}$  pore size and 52 % porosity in the sample containing 40 % sawdust. For rice husks, the values were slightly lower (9.93  $\mu\text{m}$  pore size, 47 % porosity) compared to the same concentration of sawdust with the exception of the median pore size at 20 % where the sample containing sawdust had a lower median pore size (1.75  $\mu\text{m}$  pore size, 44 % porosity) though porosity was about the same as for the sample containing rice husks (3.45  $\mu\text{m}$  pore size, 43 % porosity). For both, sawdust and rice husks the increase in median pore size was steeper than the increase in porosity.



**Figure 22:** Median pore size and porosity of clay mixtures composing of 0, 20 and 40 % sawdust (SD) or rice husks (RH).

**Pore size distribution.** To get more information about the distribution of the pore sizes, especially the existence of huge pores where bacteria can easily slip through, mercury porosimetry was conducted. The resulting pore size distribution is shown exemplarily in Figure 23 for one sample of each mixture. In general, the mixture containing no combustible material showed pores between 0.006 and 0.04  $\mu\text{m}$  and 2 to 20  $\mu\text{m}$ . With increasing concentration of sawdust or rice husks, this distribution shifted towards bigger pore sizes. Though the incremental intrusion of the samples containing rice husks was significantly lower compared to the samples containing sawdust, the pore diameters were higher. Thus, the samples containing sawdust showed a group of peaks reaching from 2 to 40  $\mu\text{m}$  pore diameter with a maximum incremental intrusion of about 0.036 mL/g at 26.8  $\mu\text{m}$  pore size for 40 % sawdust. In contrast, samples containing rice husks showed a group of peaks reaching from 2 to 100  $\mu\text{m}$  pore size (or even higher) with a maximum incremental intrusion of 0.02 mL/g at 36  $\mu\text{m}$  for 40 % rice husks.



**Figure 23:** Pore size distribution of clay mixtures composing of 0, 20 and 40 % sawdust (SD) or rice husks (RH).

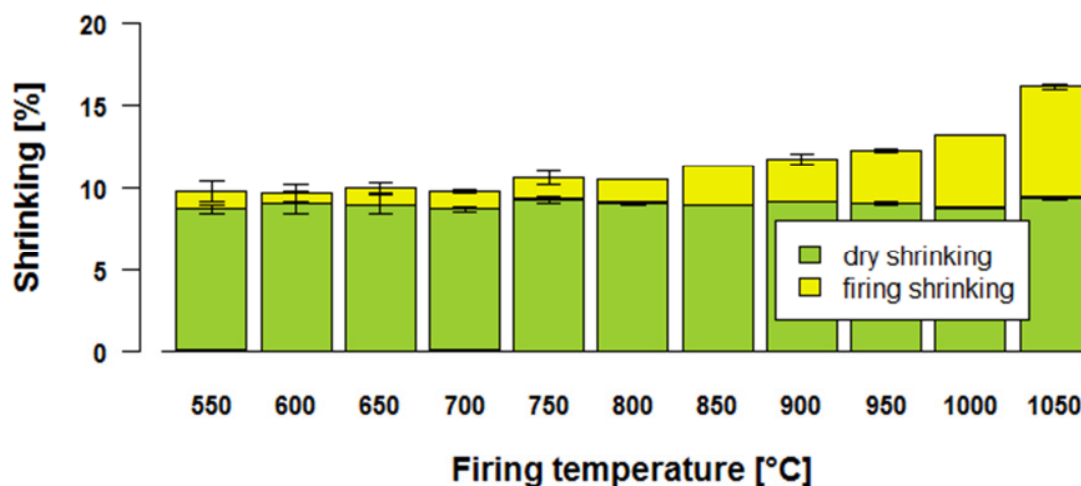
**Silver.** The pot containing 20 % sawdust that was submerged in 1.26 g/L silver nitrate for 1 min soaked 169.5 g of the solution.

### 3.3.4 Development of an appropriate firing procedure

To develop a firing procedure that is appropriate for local conditions, in addition to analyzing local firing habits, the local clay mixture was analyzed for melting behavior. Moreover, it was fired at different temperatures and the resulting ceramic plates were analyzed for differences in shrinking, density, water absorption and porosity. The results are shown in the following paragraphs.

**Melting behavior.** It could be observed that sintering occurred at a temperature of about 864 °C. Melting started at about 1400 °C.

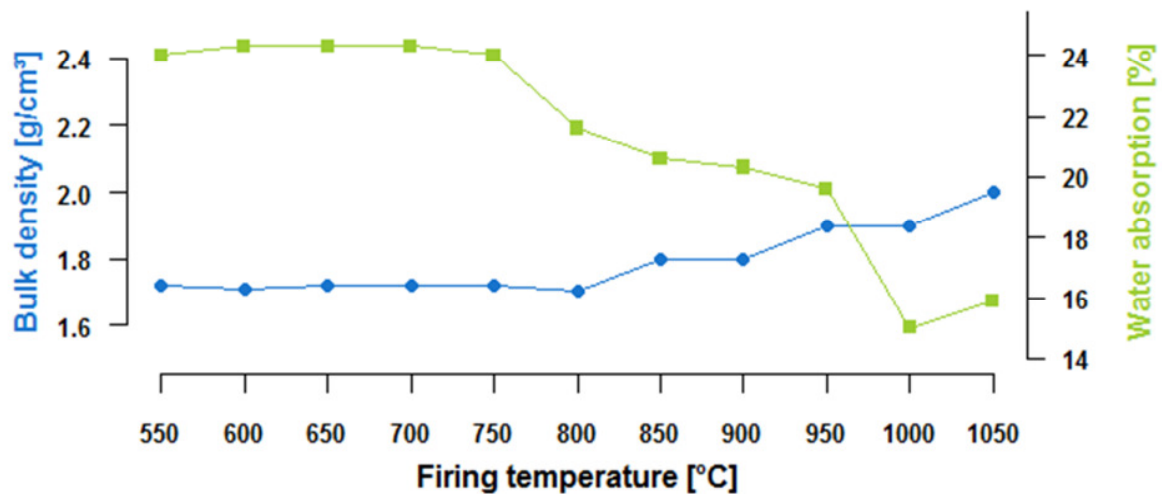
**Shrinking.** While shrinking during drying of the plates was relatively constant with 8.7 to 9.4 % (see Figure 24), the shrinking during firing rose with rising firing temperature from 1 % at 550 °C to 6.7 % at 1050 °C.



**Figure 24:** Dry and firing shrinking of traditional clay mixture at different firing temperatures.

**Bulk density and water absorption.** While bulk density of the ceramic plates was stable (1.7 g/cm<sup>3</sup>) at firing temperatures from 550 to 800 °C, the density rose when firing temperature was further increased and reached 2 g/cm<sup>3</sup> at a firing temperature of 1050 °C (see Figure 25).

Water absorption was relatively stable for ceramic plates fired at 550 to 750 °C with about 24 % and then dropped to about 20 % for plates fired at 950 °C. For plates fired at 1000 and 1050 °C, the water absorption rapidly dropped below 16 %. However, it has to be mentioned that the standard deviation being about 0.1 to 0.6 % for the other samples was 4.2 and 5.0 % for the samples fired at 1000 and 1050 °C.

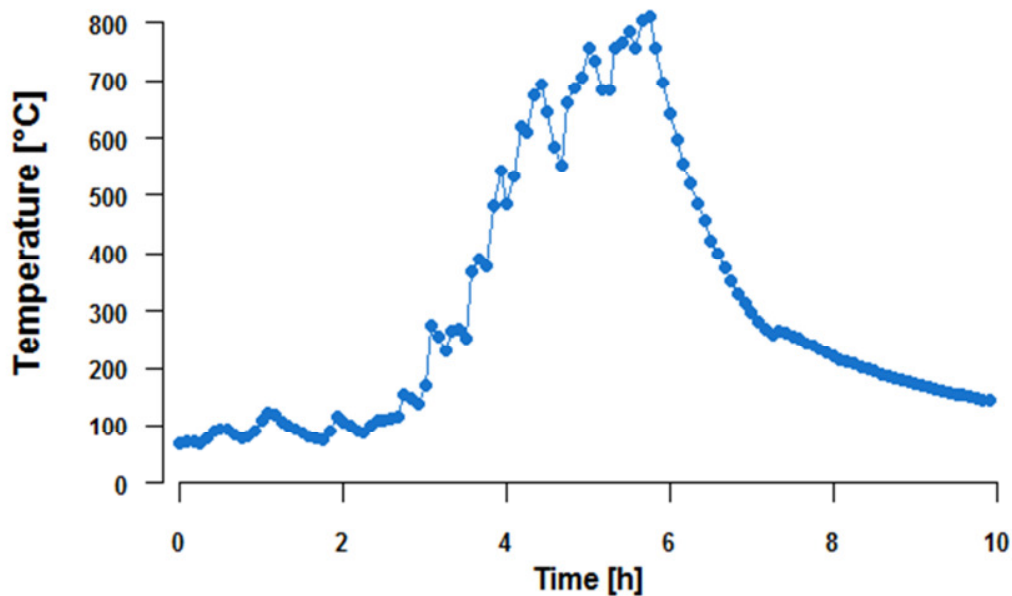


**Figure 25:** Bulk density and water absorption of traditional clay mixture at different firing temperatures.

**Higher firing temperature in local kilns.** To estimate the possibilities of local potters to fire the filter pots at higher temperatures with their existing kilns and material, the potters were asked to achieve a firing temperature of more than 800 °C.

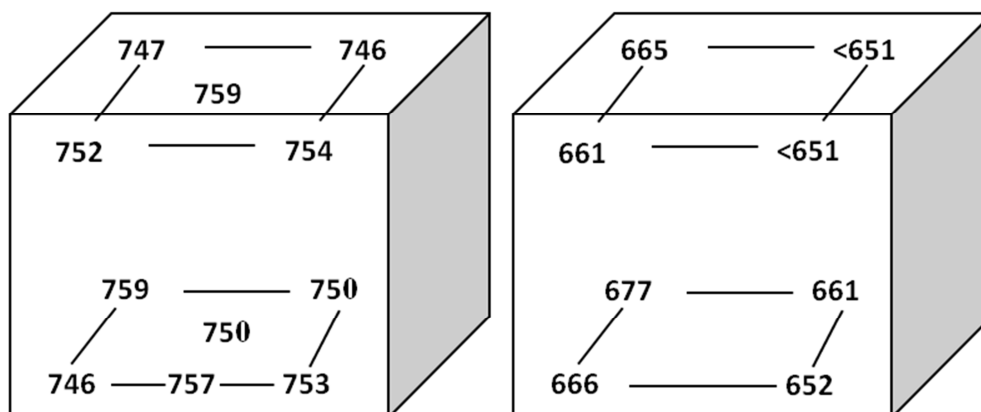
Figure 26 shows the temperature profile measured during this firing event. Similar to previous measurements, the temperature increased from 137.4 °C to 691.4 °C in 90 min resulting in a heating rate of about 6.2 K/min. Then, the temperature decreased to 552 °C before it increased again to 756.6 °C. A further decrease to 684 °C was followed by a final increase to a maximum of 811.4 °C. To increase the temperature from 552 to 811 °C it took 65 min. The peak temperature of <800 °C was held for about 10 min.





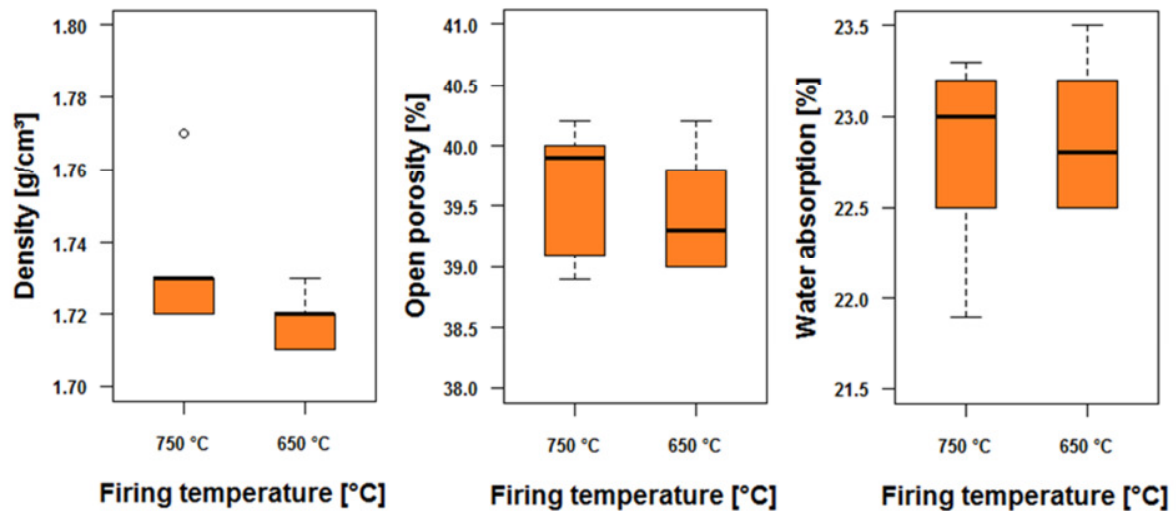
**Figure 26:** Temperature profile of the firing process aiming at 800 °C.

To evaluate the influence of the higher firing temperature compared to the traditionally achieved firing temperature in the local kilns, 10 ceramic pots were produced from one single clay mixture and 5 pots were fired at the above mentioned higher firing temperature, while the remaining 5 pots were fired at the traditional firing temperature. Figure 27 shows the distribution of the highest temperatures achieved during the firing events throughout the kiln. In general, the maximum temperature was relatively homogeneously distributed with about 746 to 759 °C in the firing event achieving a higher firing temperature and up to 677 °C in the traditional firing event.



**Figure 27:** Distribution of maximum temperature throughout the kiln during two firing events. 650 °C was the lower detection limit for the temperature control rings used in the experiments.

In Figure 28, density, porosity and water absorption of the ceramic pots fired at the different temperatures are shown. In all parameters, the ceramic fired at lower temperature showed slightly lower values. Median density was about 1.72 g/cm<sup>3</sup> for firing at 650 °C and 1.73 g/cm<sup>3</sup> for firing at 750 °C. The difference in median open porosity was about 0.6 % and the difference in median water absorption was about 0.2 %.

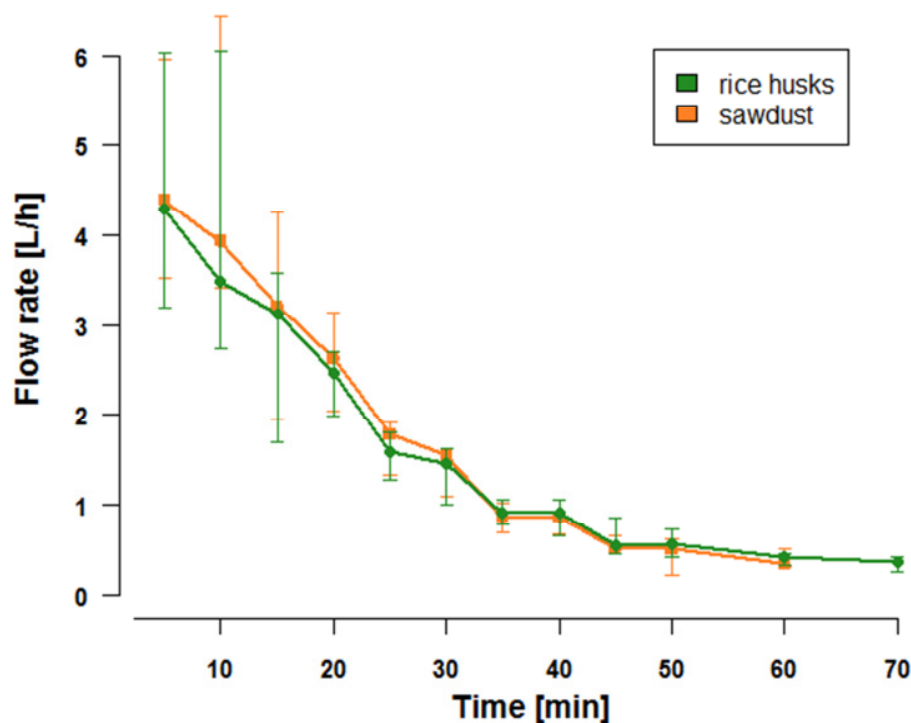


**Figure 28:** Density, open porosity and water absorption of samples fired at different temperatures.

### 3.3.5 Filter performance and development of an optimized design

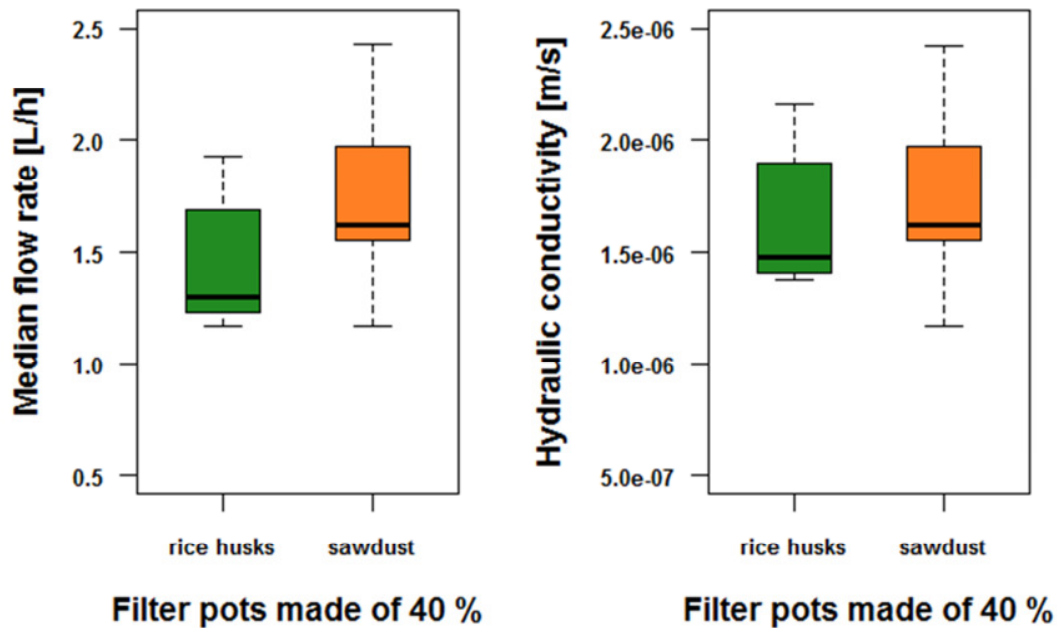
**Flow rate and hydraulic conductivity.** Flow rate is an important parameter for evaluating the filter performance. Thus, flow rates were determined depending on nature and quantity of the pore-forming agents. Following the equations given in chapter 1.8, the hydraulic conductivities of the filter pots were calculated.

First, a flow rate analysis was conducted with 9 runs each for unused filters containing 40 % rice husks or sawdust. The flow rates decreased with decreasing water head for all filters. In Figure 29, flow rates are shown exemplarily for the 9 runs of one filter pot made of rice husks and one made of sawdust.



**Figure 29:** Change in flow rate of filter pots containing 40 % sawdust or rice husks as pore-forming agents.

Throughout the 9 runs, a median flow rate of 1.2 to 1.9 L/h for the filter made of 40 % rice husks and 1.2 to 2.4 L/h for the filter containing 40 % sawdust were obtained (Figure 30). Thus, hydraulic conductivities were about  $1.4$  to  $2.1 \cdot 10^{-6}$  for rice husk filters and  $1.2$  to  $2.4 \cdot 10^{-6}$  m/s for sawdust filters.



**Figure 30:** Median flow rate (left) and corresponding hydraulic conductivity (right) of filter pots containing 40 % sawdust or rice husks as pore-forming agents.

Additionally, used filters of 0, 20 and 40 % pore-forming agents were analyzed. In Table 11, dimensions, flow rate and hydraulic conductivity of the filter pots containing rice husks and sawdust in different concentrations are given.

While the median flow rate of the filter containing no pore-forming agents was about 0.07 L/h (hydraulic conductivity  $6.8 \cdot 10^{-8}$  m/s), filters made of 20 % rice husks or sawdust had flow rates of 0.15 to 0.20 L/h (hydraulic conductivities  $1.9$  to  $2.1 \cdot 10^{-7}$  m/s). Filters made of 40 % combustibles showed flow rates of 0.29 (hydraulic conductivity  $2.6 \cdot 10^{-7}$ ) for sawdust and 0.86 L/h (hydraulic conductivity  $1.1 \cdot 10^{-6}$  m/s) for rice husks.

**Table 11:** Dimensions (see Figure 9), volume filtered so far, medium flow rate and hydraulic conductivity of the filter pots.

	rice husks			sawdust		
	0 %	20 %	40 %	20 %	40 %	20 % Ag
$r_0$ [cm]	4.85	4.9	4.55	4.8	5.15	5.2
$t_b$ [cm]	1.1	1.2	1.2	1.2	1.4	1.0
$t_s$ [cm]	1.1	1.25	1.0	1.0	1.0	1.1
$\tan\theta$ [-]	0.25	0.19	0.20	0.26	0.22	0.24
$h$ [cm]	11.3	10.5	10.0	9.7	11.3	10.2
$V$ [L]	40	44	50	47	83	45
$Q$ [L/h]	0.07	0.17	0.86	0.15	0.29	0.20
$k_f$ [m/s]	$6.8 \cdot 10^{-8}$	$2.1 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$1.9 \cdot 10^{-7}$	$2.6 \cdot 10^{-7}$	$2.1 \cdot 10^{-7}$

**Physico-chemical performance.** Physico-chemical analysis of the filtrates obtained from the filter pot containing 40 % sawdust (see Table 12) showed that the initial flow rate dropped from 1 L/h to 0.6 L/h after 8 liters were filtered. However, then it remained constant throughout the experiments. The pH of the filtrates seemed slightly increased with values varying from 8.2 to 8.6 for filtrates F2 to F18 while the pH of the feed tap water was about 7.4. Apart from an increased conductivity in the first filtrate, no significant changes in EC or hardness could be observed in the filtered water. Values for TOC behaved similarly with a peak of 5.3 mg/L in the first filtrate and varying values of 0.5 to 1 mg/L in the following filtrates (tap water 0.6 mg/L). Calcium and magnesium concentrations were not significantly influenced by filtration. However, sodium and potassium showed increased concentrations in the first filtrates decreasing again to the levels present in the tap water after 3 L (for sodium) to 8 L (for potassium) were filtered. Values for turbidity and color absorption were increased in the filtrates. The first filtrate had a turbidity peak of about 3.2 NTU (tap water 0.15 NTU) and a color absorption of 62.5 (tap water 0.9) at a wave-length of 254 nm. In the second filtrate, these values already dropped to 0.8 NTU and color absorption of 23.5 at 254 nm. Turbidity values varied from 0.18 to 0.38 NTU in filtrates F12 to F18 while absorption at 254 nm continuously decreased further to 5.4 in F18. Regarding leaching of metal elements, only arsenic and manganese showed increased concentrations in the filtrates. Both elements showed a peak in concentration in the second filtrate. Values for arsenic

decreased from 0.036 mg/L in F2 to 0.015 mg/L in F18 (0.004 mg/L in F50, data of brook water experiment). Concentrations for manganese rapidly dropped from 0.18 mg/L in F2 to below 0.01 mg/L in F14. Calcite solubility decreased from -4.7 (F1) to -67.5 mg/L (F16) while the tap water showed a value of -17 mg/L.

**Table 12:** Selected data for physico-chemical analysis of tap water (TW) and filtrates (F) of the filter pot containing 40 % sawdust.

parameter	TW	F1	F2	F3+4	F5+6	F8	F10	F12	F14	F16	F18
Flow rate [L/h]	-	-	1	-	-	0.6	0.6	0.6	-	0.5	0.6
pH [-]	7.39	7.75	8.24	8.23	8.27	8.32	8.32	8.33	8.17	8.56	8.35
El. Cond. [ $\mu$ S/cm]	716	873	662	629	638	666	688	691	705	729	701
Turbidity [NTU]	0.15	3.2	0.76	0.8	0.35	0.79	0.54	0.18	0.24	0.18	0.38
Hardness [mmol/L CaCO <sub>3</sub> ]	3.4	3.19	2.64	2.83	2.82	3.12	3.31	3.37	3.34	3.71	3.37
As [mg/L]	-	0.0006	0.036	0.024	0.022	0.021	0.02	0.019	0.021	0.021	0.015
Mn [mg/L]	<0.005	< 0.01	0.18	0.15	0.14	0.091	0.031	0.016	< 0.01	< 0.01	< 0.01
Na [mg/L]	16.6	30.7	19.3	15.8	15.9	16.6	16.3	16.7	16.3	16.7	16.4
K [mg/L]	3.5	21.8	10.2	8.55	4.69	3.8	3.32	3.33	3.19	3.28	3.21
Mg [mg/L]	14.7	12.1	10.6	11.4	12.2	13.2	13.6	13.9	14	15.2	14.1
Ca [mg/L]	112	107.7	96.2	93.1	93	103.1	110.5	111.9	112.4	123.4	111.1
Calcite solubility [mg/L]	-17	-4.73	-24.86	-29.63	-31.38	-42.91	-49.24	-51.16	-47.14	-67.54	-53.22
TOC [mg/L]	0.56	5.3	0.76	0.78	1.1	0.68	0.77	0.47	0.71	0.69	0.69
color (absorption at 436 nm)	<0.1	0.33	0.15	0.08	0.07	0.06	0.06	0.06	0.06	0.08	0.06
color (absorption at 254 nm)	0.9	62.5	23.5	14.4	13.9	13.1	11.6	9.6	8.8	7.2	5.4

The second filtrate (F2) of the filter containing 40 % rice husks showed similar values compared to F2 of the filter made of 40 % sawdust with a concentration of 0.012 mg/L arsenic and 0.47 mg/L manganese (see Table 13). Arsenic concentration dropped to 0.006 mg/L in F7 and 0.003 mg/L in F10 while manganese concentration dropped to 0.048 mg/L in F7.

Following this analyses of the filters made of 40 % sawdust or rice husks, filters containing 20 % or no combustible material were analyzed concerning their performance. Moreover, the effect of a silver treatment was determined. Therefore, a filter made of 20 % sawdust that was treated with silver nitrate was included in the experiment setup. Selected data for the second filtrates F2 are shown in Table 13. While the initial flow rate of the filter containing 40 % rice husks was about 1.5 L/h, the flow rate for the filters containing 20 % rice husks or sawdust was about 0.2 to 0.3 L/h. The filter containing no additional combustible material had a flow rate of about 0.15 L/h. Calcite solubility was about -43.3 mg/L for the filtrate of the filter made of 40 % rice husks, -19.3 to -23.4 for filters made of 20 % sawdust and 0.4 mg/L for the filtrate of the filter without rice husks. Arsenic concentration decreased from 0.013 (F1, 40 % R) and 0.038 (F1, 20 % R) to 0.006 (F7, 40 % R) and 0.008 mg/L (F7, 20 % R) for rice husk filters. Filters containing 20 % sawdust leached arsenic in concentrations of 0.008 to 0.013 mg/L in filtrates F5 and F10. For the pot without rice husks, concentrations varied between 0.001 (F2, 0 %) and 0.006 mg/L (F7, 0 %) (complete data in appendix). For manganese, values dropped from 0.47 (40 % rice husks), 0.44 (20 % rice husks) and 0.72 mg/L (no rice husks) in F2 to 0.048 (40 % rice husks), 0.040 (20 % rice husks) and 0.033 mg/L (no rice husks) in F7. For sawdust filters, manganese values were higher in the 5<sup>th</sup> filtrate F5 with 0.08 to 0.1 mg/L with very low concentrations in the 10<sup>th</sup> filtrate (<0.01 to 0.02 mg/L). Moreover, it was visible that silver was leached from the coated filter with a concentration of 0.014 mg/L. This concentration was about 0.024 in the 5<sup>th</sup> filtrate and dropped to 0.018 mg/L in the 10<sup>th</sup> filtrate (data not shown). After 22 liters were filtered, this concentration dropped to below 0.003 mg/L in the brook water experiments.



**Table 13:** Selected data for physico-chemical performance of tap water (TW) and second filtrates F2 (F) of filter pots containing rice husks (R) and sawdust (S) in different concentrations.

F20 Ag = filtrate of filter made of 20 % sawdust coated with silver nitrate.

parameter	TW	F40 R	F20 R	F0	F20 S	F20 Ag
Flow rate [L/h]	-	1.5	0.2	0.15	0.2	0.3
As [mg/L]	-	0.012	0.013	0.001	0.011	0.005
Mn [mg/L]	<0.005	0.47	0.44	0.72	<0.01	<0.01
Ag [mg/L]	<0.003	-	-	-	<0.003	0.014
Calcite solubility [mg/L]	-17	-43.3	-	0.444	-19.3	-23.4

Subsequently, experiments were conducted applying natural water from a river and brook to the filter pots. To determine the effect of a fractioned filtration, water was first fed to the ceramic pot made of 40 % rice husks. The filtrate was then applied to the 20 % pot and the resulting filtrate was applied to the pot without rice husks. Results of physico-chemical analysis are shown in Table 14.

While the brook water had a turbidity of 27.6 NTU and an EC of 645  $\mu\text{S}/\text{cm}$ , the river water had a turbidity of 3.7 NTU and an EC of 367  $\mu\text{S}/\text{cm}$ . The initial flow rate of the brook water through the first filter (40 % R) was half as high as for river water. While turbidity was decreased to 0.1 to 0.16 NTU through filtration, EC was slightly decreased for the brook water with high conductivity values and slightly increased for the river water with low conductivity values. Hardness was not changed significantly through filtration. While arsenic was still leached from the filters in very low concentrations (0.006 mg/L was the highest value detected), no increased values for manganese could be detected (<0.01 mg/L). Values for calcite solubility and concentration of sodium, potassium, magnesium and calcium varied in the filtrates. TOC was slightly decreased through filtration. TOC values decreased from 1 mg/L in the brook water to 0.79 mg/L in the third filtrate and from 1.7 mg/L in river water to 1.3 mg/L in the third filtrate. Color absorption at 436 nm decreased slightly from 0.18 in brook water and 0.27 in river water to 0.05 and 0.12 in the filtrates of the fractioned filtration (40+20+0 %). In contrast, values for color absorption at 254 nm were slightly increased in the filtrates.

**Table 14:** Selected data for physico-chemical analysis of natural brook and river water filtered through rice husk filter pots. BW = brook water, RW = river water, F40 = filtered through filter containing 40 % rice husks, F40+20 = F40 subsequently filtered through filter containing 20 % rice husks, F40+20+0 = F20 filtered through filter containing no rice husks.

Parameter	Brook water				River water			
	BW	F40	F40+20	F40+20+0	RW	F40	F40+20	F40+20+0
Flow rate [L/h]	-	0.5	0.25	0.09	-	1.34	0.16	0.05
pH [-]	7.94	8.53	8.57	8.09	7.78	8.06	8.19	8.26
El. Cond. [ $\mu\text{S}/\text{cm}$ ]	645	629	618	556	367	373	383	428
Turbidity [NTU]	27.6	0.23	0.21	0.19	3.7	0.26	0.24	0.30
Hardness [mmol/L $\text{CaCO}_3$ ]	3.27	3.25	3.18	2.6	1.58	1.63	1.63	1.95
As [mg/L]	0.002	0.004	0.006	0.003	<0.0005	0.001	0.002	0.006
Mn [mg/L]	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Na [mg/L]	9.88	10.2	9.77	12	11.8	11.9	12.3	13.5
K [mg/L]	1.93	1.9	1.89	2.6	2.37	2.25	2.13	1.87
Mg [mg/L]	19.9	19.3	16.2	13.1	7.56	7.84	8.82	10.7
Ca [mg/L]	98.3	98.2	100.8	82.6	50.8	52.4	50.7	60.4
Calcit solubility [mg/L]	-42.1	-5.2	-55.9	-24.9	-2	-7.7	-9.8	-15.1
TOC [mg/L]	1	0.95	0.86	0.79	1.7	1.4	1.4	1.3
color (absorption at 436 nm)	0.18	0.13	0.09	0.05	0.27	0.19	0.17	0.12
color (absorption at 254 nm)	2.68	3.81	4.06	3.66	4.79	4.76	5.04	6.19

The brook water experiment was repeated with filters made of 40 and 20 % sawdust and a 20 % sawdust filter which was coated with silver nitrate. Selected data for physico-chemical performance are shown in Table 15. In general, they resembled the rice husk filter results.

**Table 15:** Selected data for physico-chemical analysis of natural brook water filtered through pots.

BW = brook water, F20 S = filtered through filter containing 20 % sawdust, F20 Ag = filtered through filter containing 20 % sawdust and coated with silver nitrate, F40 S = filtered through filter containing 40 % sawdust.

parameter	BW	F40 S	F20 S	F20 Ag
Flow rate [L/h]	-	0.4	0.4	0.4
pH [-]	8.04	8.22	8.04	8.29
El. Cond. [ $\mu\text{S}/\text{cm}$ ]	774	772	776	776
Turbidity [NTU]	9.6	0.26	0.34	0.1
Hardness [m mol/L $\text{CaCO}_3$ ]	3.83	3.73	3.67	3.74
As [mg/L]	0.002	0.004	0.005	0.012
Mn [mg/L]	0.02	< 0.01	< 0.01	0.1
Ag [mg/L]	<0.003	<0.003	<0.003	<0.003
Na [mg/L]	14.2	16.2	16.8	16.0
K [mg/L]	2.7	3.57	5.56	4.49
Mg [mg/L]	23.0	20.6	20.6	19.0
Ca [mg/L]	115.5	115.4	113.1	118.3
Calcite solubility [mg/L]	-69.8	-72.4	-58.3	-67.2
TOC [mg/L]	2.1	2.7	4.2	5.5
color (absorption at 436 nm)	0.17	0.29	0.25	0.2
color (absorption at 254 nm)	5.7	5.4	10.4	6.69

**Microbiological performance.** In addition to physico-chemical performance analysis, microbiological performance was analyzed in the filtration experiments with brook and river water. It was remarkable that heterotrophic plate count (HPC) was higher in the filtrates compared to the raw water, while coliform bacteria and enterococci were reduced through filtration. As an example, values are given in Table 16 for the first brook water experiments for rice husk filters (complete data in appendix). The only exception

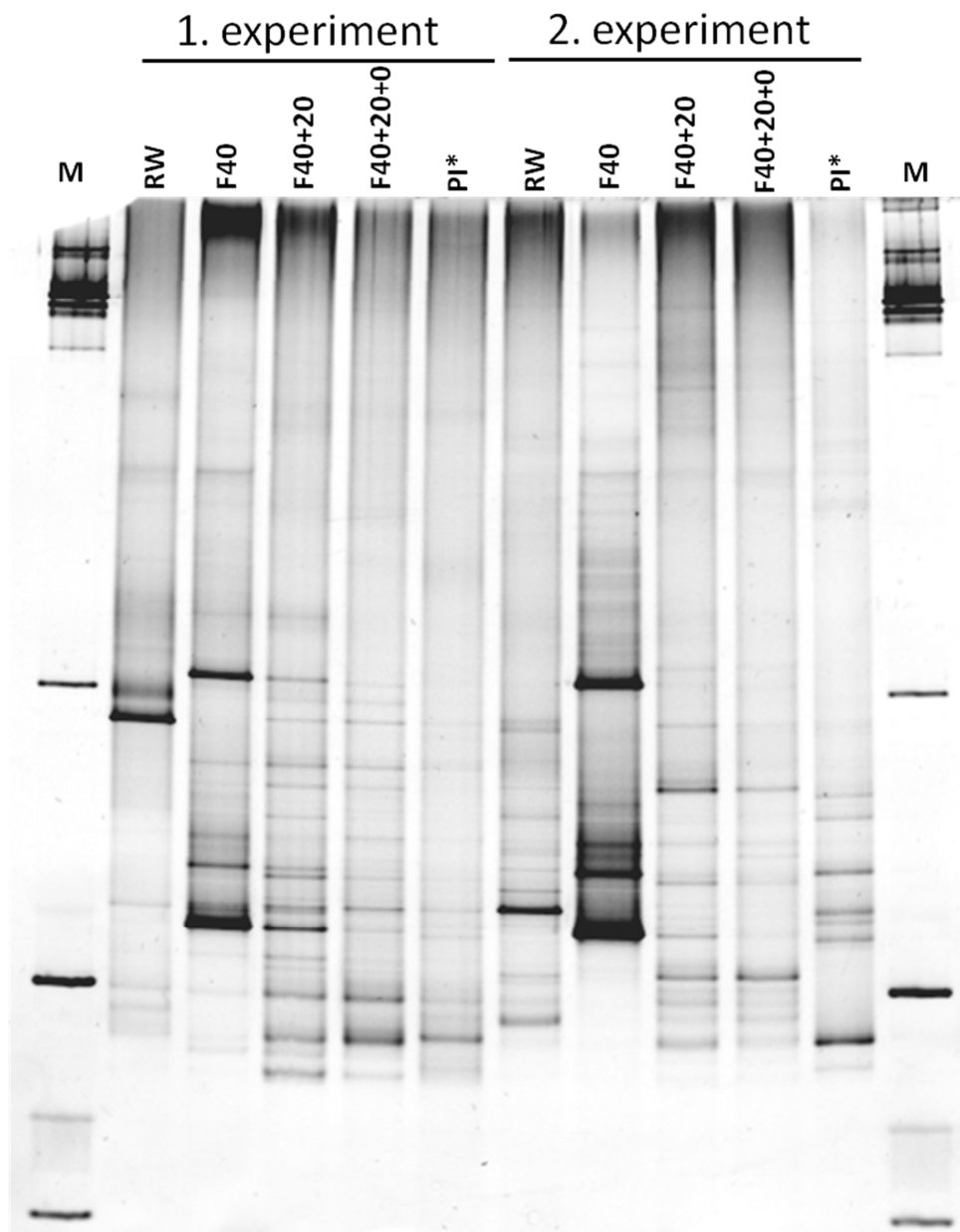
was the filter treated with silver nitrate whose filtrate showed only 1 to 70 CFU/mL at 36 °C and 2 to 2,000 CFU/mL at 22 °C. However, results showed that with repetition of the experiments the HPC counts increased.

**Table 16:** Values for heterotrophic plate count (HPC), presence/absence of coliform bacteria and enterococci for the first brook water experiment

BW = brook water, F40 R = BW filtered through filter with 40 %rice husks, F40+20 R = F40 R filtered through filter with 20 % rice husks, F40+20+0 R = F40+20 R filtered through filter containing no rice husks.

parameter	BW	F40 R	F40+20 R	F40+20+0 R
HPC at 22 °C [CFU/mL]	$6 \cdot 10^3$	$3.6 \cdot 10^4$	$2.8 \cdot 10^4$	$2.1 \cdot 10^5$
HPC at 36 °C [CFU/mL]	$1.2 \cdot 10^3$	$1.3 \cdot 10^4$	$1.8 \cdot 10^4$	$4.4 \cdot 10^4$
Coliform bacteria [CFU/100 mL]	+++	++	++	+
Enterococci [CFU/100 mL]	+++	++	+	+

Population analysis using DGGE showed that the band pattern varied within the filtration course. In Figure 31, the band pattern of the river water experiments is given exemplarily for the rice husk filters. It is remarkable that even the band pattern of the raw water varied from the two samples taken of the brook on the same day. In the filtrates, different bands appeared. While the pattern of the 40 % filtrate showed only few but dominant bands, the pattern of the water filtered through 40 and 20 % filters (F40+20) and subsequent 0 % filters (F40+20+0) showed quite a lot of less dominant bands. The overall band pattern of F40+20 to F40+20+0 was quite similar.



**Figure 31:** DGGE pattern of river water samples.

RW = river water, F40 = RW filtered through filter with 40 %rice husks, F40+20 = F40 filtered through filter with 20 % rice husks, F40+20+0 = F20 filtered through filter containing no rice husks.\*R PI represents the filtrate of the ceramic filter produced by *Pelita*.

Sequencing revealed that bacteria mostly belonged to beta and gamma proteobacteria (see Table 17). In brook water, there were only beta proteobacteria, while in river water cyanobacteria, firmicutes and actinobacteria were detected. Bacteria that could only be detected in the filtered water, such as *Methylobacterium*, *Acidovorax*, *Limnobacter*, *Limnohabitans* and *Acinetobacter*, mostly belonged to the proteobacteria groups.

**Table 17:** Bacteria sequencing results from DGGE gels for brook and river water experiments with rice husk filters.

BW = brook water, F40 = BW filtered through filter with 40 %rice husks, F40+20 = F40 filtered through filter with 20 % rice husks, F40+20+0 = F20 filtered through filter containing no rice husks.

+ = band was clearly visible. (+) = band was very thin.

bacterium	BW	F40	F40+20	F40+20+0	RW	F40	F40+20	F40+20+0
<b>Alpha proteobacteria</b>								
Gemmobacter sp.							+	
Methylobacterium sp.			+				+	
<b>Beta proteobacteria</b>								
Acidovorax sp.		+	+					
Ferribacterium sp.	+							
Limnobacter sp.		+	+			+	+	
Limnohabitans sp.		+	+	+		+	+	
Undibacterium sp.	+							
Zoogloea sp.	(+)	+	+	+			+	+
<b>Gamma proteobact.</b>								
Acinetobacter sp.		+	+			+	+	+
Nevskia sp.		+						
Rheinheimera sp.				+				
<b>Cyanobacteria</b>								
Hydrocoleum sp.					+			
<b>Firmicutes, Bacilli</b>								
Exiguobacterium sp.					+			+
<b>Actinobacteria</b>								
Actinobacterium sp.				+	+	+	+	+
Nocardioides sp.		+	+			+	+	

To get a more detailed insight in log reduction potential for pathogen bacteria, tap water spiked with *E. coli*, *Ent. faecium* and *P. aeruginosa* was applied to the filter pots.

Filtering through a filter made of 40 % rice husks achieved a log reduction of about 0.6 to 0.7 for *E. coli*, 0.5 for *Ent. faecium* and 1.1 to 1.2 for *P. aeruginosa*. This log reduction increased with supplementary filtering steps. The log reduction after filtering through the

40 %, 20 % and 0 % filter was about 1.7 to 1.9 for *E. coli*, 3.1 to more than 3.7 for *Ent. faecium* and 2.4 to more than 4.5 for *P. aeruginosa* (Table 18).

**Table 18:** Bacteria count [CFU/mL] and <sup>8</sup>log removal for *E. coli*, *Ent. faecium* and *P. aeruginosa*.

RAW = raw water (spiked tap water), F40 = RAW filtered through filter with 40 %rice husks, F40+20 = F40 filtered through filter with 20 % rice husks, F40+20+0 = F20 filtered through filter containing no rice husks.

		First experiment			Repetition		
		<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeruginosa</i>	<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeruginosa</i>
Bacteria count [CFU/mL]	RAW	1.7*10 <sup>7</sup>	1.1*10 <sup>6</sup>	3.1*10 <sup>6</sup>	1.2*10 <sup>7</sup>	5.5*10 <sup>5</sup>	2.7*10 <sup>6</sup>
	F40	3.8*10 <sup>6</sup>	3.8*10 <sup>5</sup>	1.9*10 <sup>5</sup>	3.2*10 <sup>6</sup>	1.9*10 <sup>5</sup>	2.2*10 <sup>5</sup>
	F40+20	8.3*10 <sup>5</sup>	3.8*10 <sup>4</sup>	6.1*10 <sup>3</sup>	2*10 <sup>6</sup>	3.5*10 <sup>4</sup>	1*10 <sup>5</sup>
	F40+20+0	2.1*10 <sup>5</sup>	8*10 <sup>2</sup>	<1*10 <sup>2</sup>	2.4*10 <sup>5</sup>	<1*10 <sup>2</sup>	1.2*10 <sup>4</sup>
Log reduction [-]	F40	0.7	0.5	1.2	0.6	0.5	1.1
	F40+20	1.3	1.5	2.7	0.8	1.2	1.4
	F40+20+0	1.9	3.1	>4.5	1.7	>3.7	2.4

Quantitative PCR results showed a log removal of 0 to 1 for *E. coli*, 0.3 to 0.7 for *Ent. faecium* and 0 to 0.3 for *P. aeruginosa* for the filter made of 40 % rice husks. A combination of 40 % and 20 % rice husk filters showed a log removal of 0.9 to 1.5 for *E. coli*, 1.7 to 1.8 for *Ent. faecium* and 0.7 for *P. aeruginosa*. Fractioned filtration through 40 %, 20 % and 0 % rice husks filters showed a log removal of 1.6 to 2.2 for *E. coli*, 3.3 to 4.8 for *Ent. faecium* and 1.2 to 2.0 for *P. aeruginosa*. Thus, an increased log reduction with additional filtering steps could be confirmed.

Bacteria count results for the sawdust filters are shown in Table 19. In addition to fractioned filtration, the filters were also tested individually. It was remarkable that, in this case, a fractioned filtration showed no increased log reduction compared to single filtration through the filter made of 20 % or 0 % combustibles. In general, filtration through the silver-treated filter had the highest log reduction with more than 4 to 5 log steps for all tested bacteria followed by the 0 % filter with 1.8 to 2.5 log steps for *E. coli*,

<sup>8</sup> Log removal = log<sub>10</sub> (bacteria count in RAW / bacteria count in F)

1.9 to 2.4 log steps for *Ent. faecium* and 2.6 to 3.2 log steps for *P. aeruginosa*. In these experiments, silver leached from the filter was below a concentration of 0.003 mg/L.

**Table 19:** Bacteria count [CFU/mL] and log removal for *E. coli*, *Ent. faecium* and *P. aeruginosa*.

RAW = raw water (spiked tap water), F40 = RAW filtered through filter with 40 % sawdust, F20 = RAW filtered through filter with 20 % sawdust, F0 = RAW filtered through filter containing no sawdust, F40+20 = F40 filtered through filter with 20 % sawdust, F40+20+0 = F40+20 filtered through filter containing no sawdust, F20 Ag = RAW filtered through filter with 20 % sawdust and treated with silver nitrate, F40+20 Ag = F40 filtered through filter with 20 % sawdust and treated with silver nitrate.

	First experiment			Repetition			
	<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeru- ginosa</i>	<i>E. coli</i>	<i>Ent. faecium</i>	<i>P. aeru- ginosa</i>	
Bacteria count [CFU/mL]	RAW	4.9*10 <sup>5</sup>	4.8*10 <sup>5</sup>	1.6*10 <sup>5</sup>	2.4*10 <sup>6</sup>	1.5*10 <sup>6</sup>	1.8*10 <sup>6</sup>
	F40	7.1*10 <sup>5</sup>	1.4*10 <sup>5</sup>	1.2*10 <sup>5</sup>	2.1*10 <sup>5</sup>	3.9*10 <sup>5</sup>	1.8*10 <sup>4</sup>
	F20	4.0*10 <sup>2</sup>	7.0*10 <sup>3</sup>	6.0*10 <sup>2</sup>	7.0*10 <sup>4</sup>	7.9*10 <sup>4</sup>	2.4*10 <sup>3</sup>
	F0	1.7*10 <sup>3</sup>	2.0*10 <sup>3</sup>	1.1*10 <sup>2</sup>	3.7*10 <sup>4</sup>	1.7*10 <sup>4</sup>	4.9*10 <sup>2</sup>
	F40+20	3.5*10 <sup>3</sup>	9.5*10 <sup>3</sup>	1.3*10 <sup>3</sup>	1.4*10 <sup>5</sup>	6.8*10 <sup>4</sup>	1.7*10 <sup>4</sup>
	F40+20+0	6.2*10 <sup>4</sup>	2.8*10 <sup>4</sup>	4.6*10 <sup>3</sup>	9.6*10 <sup>4</sup>	2.8*10 <sup>4</sup>	8.1*10 <sup>3</sup>
	F20 Ag	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	3.5*10 <sup>1</sup>	<1*10 <sup>1</sup>
	F40+20 Ag	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>	<1*10 <sup>1</sup>
Log reduction [-]	F40	0	0.5	0.1	1.1	0.6	1.0
	F20	3.1	1.8	2.4	1.5	1.3	1.9
	F0	2.5	2.4	3.2	1.8	1.9	2.6
	F40+20	2.1	1.7	2.1	1.2	1.3	1.0
	F40+20+0	0.9	1.2	1.5	1.4	1.7	1.4
	F20 Ag	>4.7	>4.7	>4.2	>5.4	4.6	>4.3
	F40+20 Ag	>4.7	>4.7	>4.2	>5.4	>5.2	>4.3

In addition to bacteria removal capacity, the filters were analyzed regarding their removal capacity for viruses. Therefore, tap water was spiked with  $\phi$ X174 phages that were used as surrogates for viruses. Results of the filter experiments are shown in Table 20. In general, achieved log reductions were up to 1 for uncoated filters and 1.2 to 1.9 log steps for silver-treated filters. Single testing of sawdust filters showed that if used



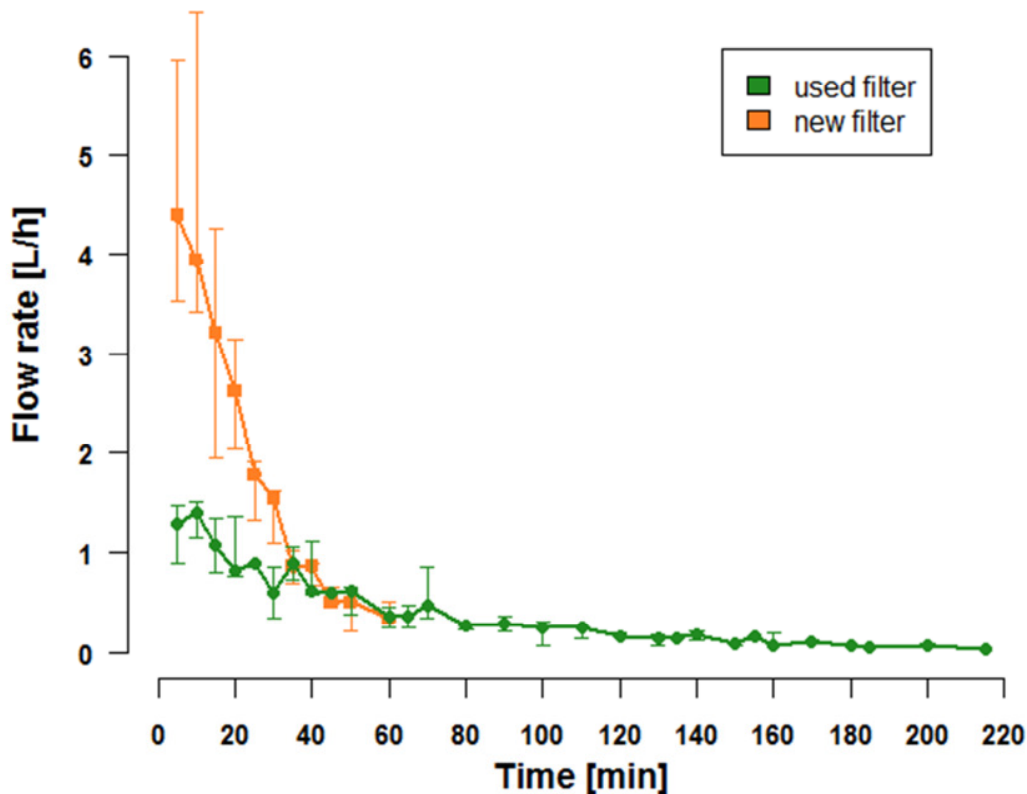
individually, the 40 % sawdust filter had the highest removal capacity of about 0.4 to 0.6 log steps. Less pore-forming agents led to decreased log reduction. Supplementary filtration steps generally increased the log reduction. However, silver-treated filters showed higher log reductions with 1.2 to 1.6 for single filtration through the 20 % sawdust filter treated with silver nitrate. If water was previously filtered through the 40 % sawdust filter, a log reduction of 1.6 to 3 log steps could be achieved for  $\phi$ X174.

**Table 20:** Phage count [PFU/mL] and log removal for  $\phi$ X174.

RAW = raw water (spiked tap water), F40 = RAW filtered through filter with 40 % sawdust (S) or rice husks (R), F20 = RAW filtered through 20 % filters, F0 = RAW filtered through 0 % filter, F40+20 = F40 filtered through 20 % filters, F40+20+0 = F40+20 filtered through 0 % filter, F20 Ag = RAW filtered through filter with 20 % sawdust and treated with silver nitrate, F40+20 Ag = F40 filtered through filter with 20 % sawdust and treated with silver nitrate.

$\phi$ X174		First experiment		Repetition	
		Phage count [PFU/mL]	Log red- uction [-]	Phage count [PFU/mL]	Log red- uction [-]
Rice husks	RAW	$7.4 \cdot 10^2$		$8.0 \cdot 10^2$	
	F40 R	$3.5 \cdot 10^2$	0.3	$3.3 \cdot 10^2$	0.4
	F40+20 R	$2.5 \cdot 10^2$	0.5	$3.5 \cdot 10^2$	0.4
	F40+20+0 R	$7.7 \cdot 10^1$	1.0	$1.3 \cdot 10^2$	0.8
Sawdust	RAW	$2.6 \cdot 10^2$		$6.4 \cdot 10^2$	
	F40 S	$6.5 \cdot 10^1$	0.6	$2.4 \cdot 10^2$	0.4
	F20 S	$2.6 \cdot 10^2$	0	$4.5 \cdot 10^2$	0.1
	F0	$2.2 \cdot 10^2$	0.1	$3.1 \cdot 10^2$	0.3
	F40+20 S	$7.7 \cdot 10^1$	0.5	$9.9 \cdot 10^1$	0.8
	F40+20+0 S	$7.5 \cdot 10^1$	0.5	$8.5 \cdot 10^1$	0.9
	F20 S Ag	7.0	1.6	$3.9 \cdot 10^1$	1.2
	F40+20 S Ag	3.0	1.9	$1.5 \cdot 10^1$	1.6

**Fouling.** As mentioned above, the flow rate of the filters decreased with usage. However, the decrease was most obvious for the filters made of 40 % combustibles. For instance, the hydraulic conductivity of the filter made of 40 % sawdust decreased from  $1.7 \cdot 10^{-6}$  (unused) to  $2.6 \cdot 10^{-7}$  m/s after 80 batches were filtered (both produced from the same mixture and fired in the same batch). Respective flow rates are given in Figure 32.



**Figure 32:** Median flow rate of a new and used filter made of 40 % sawdust.

As a sort of worst case scenario, natural brook water was filtered. The filters made of rice husks and sawdust showed similar results. Here, rice husk filter results are described exemplarily. Before applying brook water, 23 L of tap water had been filtered and the initial flow rate (first 200 mL) was about 1.5 L/h (tap water). After filtration of 18 batches of brook water, the flow rate was down to about 0.5 L/h (first 240 mL, tap water). Subsequently, the filter was cleaned with tap water and a soft brush. The flow rate after cleaning was about 1.8 L/h (first 300 mL). In Figure 33, a filter made of 40 % rice husks is shown after filtration of 18 batches of brook water with a turbidity of about 28 NTU. It is visible that a brown layer of turbid particles had formed at the bottom of the filter. However, the sides were relatively clean. After cleaning with tap water and a soft brush, most of the layer had been removed.



**Figure 33:** 40 % rice husk filter after filtering brook water (left) and after subsequent cleaning (right)

Turbidity in the filtrate of the filter made of 40 % rice husks was down to 0.23 NTU. The subsequent filters (20 % and 0 %) in the fractioned filtration did not show a significant decrease in flow rate or filtrate turbidity during brook water filtration (Table 21). Moreover, there was no layer of particles visible. Cleaning with tap water and brush increased the flow rate of all filters.

**Table 21:** Flow rate and turbidity.

\*Flow rate was measured with tap water.

parameter	BW	40 R filter	20 R filter	0 R filter
Turbidity [NTU] brook water/filtrate	27.6	0.23	0.21	0.19
*Flow rate [L/h] before brook water	-	1.5	0.2	0.1
Flow rate [L/h] with brook water	-	0.2	0.1	0.1
*Flow rate [L/h] after brook water	-	0.5	0.4	0.25
*Flow rate [L/h] after cleaning	-	1.8	0.4	0.24

### 3.4 Summary

As a basis for developing an optimized ceramic filter for POU water treatment in Gunungkidul, an existing silver-treated filter produced by *Pelita* in Western Java was analyzed. Results showed that arsenic and manganese were leached from the filter in the first batches. Log reduction for bacteria was 3 to 5 log steps while phages were reduced by 0.5 to 0.9 log steps.

In order to develop an optimized ceramic filter for use in Gunungkidul, in a next step, locally available material and traditional local firing procedures were analyzed and evaluated for production of ceramic filters.

The analysis of locally available material showed that silica, alumina and iron (III) oxide were predominant in all samples. Mineral phases included hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) and maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ), calcite ( $\text{Ca}[\text{CO}_3]$ ) and a huge group of quartz and silicates.

In the traditional local firing processes, the maximum temperature was between 520 and 750 °C. Firing shrinking, bulk density and water absorption were relatively constant at firing temperatures from 550 °C up to 800 °C.

Rice husks and sawdust as pore-forming agents were mixed with the traditional local clay mixture in varying concentrations to attain a usable filter with sufficient flow rates. With increasing concentration of sawdust or rice husks, the pore size distribution shifted towards bigger pore sizes. The samples containing rice husks had fewer big pores but the pore diameters were higher compared to the samples containing sawdust.

Unused filters made of 40 % combustibles had similar hydraulic conductivities for rice husks and sawdust with about  $1.5$  to  $2.0 \times 10^{-6}$  m/s. Used filters made of 40 % combustibles showed hydraulic conductivities of  $2.6 \times 10^{-7}$  m/s (sawdust, 80 batches filtered) to  $1.1 \times 10^{-6}$  m/s (rice husks, 50 batches filtered), filters made of 20 % combustibles had  $1.9$  to  $2.1 \times 10^{-7}$  m/s and the filter without pore-forming agents had a hydraulic conductivity of  $6.8 \times 10^{-8}$  m/s.

Similar to the filter bought in Western Java, arsenic and manganese were leached from the ceramic filter pots with concentrations up to 0.036 mg/L of arsenic and up to 0.72 mg/L of manganese in the second filtrate. These decreased with following batches. Bacteria log reduction for a fractioned filtration with rice husk filters (40+20+0 %) achieved about 1.7- 1.9 log steps for *E. coli* to 2.4- >4.5 log steps for *P. aeruginosa*. For sawdust filters, maximum log reduction was achieved for filters without combustibles

(single) with 1.8- 2.5 log steps for *E. coli* and 2.6 to 3.2 log steps for *P. aeruginosa*. For phages, log reduction was highest for fractioned filtration (40+20+0 %) with 0.8 to 1.0 log step for rice husk filters and 0.5 to 0.9 log steps for sawdust filters. Silver-treated filters achieved log reductions of about 4 to 5 log steps for bacteria and 1.2 to 1.9 log steps for phages.

## 4 Discussion

In Gunungkidul, a central supply with safe drinking water is not manageable in the next future. Therefore, POU water treatment is needed to prevent waterborne illnesses. Possible technologies comprise techniques that do not require power supply, as a reliable power supply is not given in every household. Thus, possible techniques would be solar disinfection, chemical disinfection, (bio)sand and ceramic filtration.

As discussed in chapters 1.4 and 1.5, boiling is effective against all classes of waterborne pathogens when done correctly. However, boiling as it is performed in the project region is often not done properly, barely sustainable, consumes a lot of energy (Sobsey, 2002), is labor-intensive as people in the project region need to collect wood as fuel and may contribute to respiratory diseases. Solar disinfection (SODIS) was not chosen, as its efficiency is highly dependent on sun intensity and turbidity of the water. Though sun intensity in the tropics is generally high, in the rainy season it might not be as reliable as in the dry season. Moreover, especially in the rainy season, the source water carries a lot of turbidity which reduces the SODIS efficiency. Chemical disinfection, for instance with chlorine, is known to be very efficient, but it leads to formation of by-products (Solsona and Méndez, 2003) and it requires relatively high understanding of users. Moreover, the change in taste might result in lower acceptance. Previous experiments showed that chlorination with the locally available calcium hypochlorite resulted in varying concentrations of free chlorine which was probably due to varying raw water quality and quality of used chemicals (Matthies *et al.*, 2014). To guarantee safe water, a permanent control of the disinfection effect would be needed which is not feasible in household use.

Filtration, which has the potential to reduce turbidity as well as pathogens, seems to be the most appropriate technology for use in Gunungkidul. Both, sand and ceramic filters have the potential to reduce bacteria efficiently. For the following reasons, it was decided to use ceramic filters based on those promoted by *Potters for Peace*. The increased pathogen removal efficiency for ceramic filters was only one aspect. In contrast to sand filters, ceramic filters can be transported on a motor cycle which is the most widely used vehicle in the project region. As a sand filter is built of concrete and sand which is both heavy (WHO/UNICEF, 2012), people will have to rent a car for

transport and invest quite some time for construction of the sand filter. Thus, ceramic filters are better accessible for local people. In this dissertation, a potential local production of ceramic filters was examined. Results showed that locally available raw material provides a perfect base for the production of ceramic pot filters. Moreover, the material proved to be very robust so that local firing habits with temperatures of 600 to 750 °C resulted in usable products. Filters made of local clay and 20 % combustible material that were treated with silver nitrate proved to be highly protective against bacteria and still had a sufficient flow rate to supply enough drinking water for a family of four. Field experiments proved that this kind of filter is effective under local conditions and is well accepted by local people. Last but not least, a local production of ceramic filters promotes the local economy (WHO/UNICEF, 2012). These aspects are discussed in the following paragraphs.

#### **4.1 Optimization potential for ceramic filters – analysis of an existing filter as a base for optimization**

As a base for developing a locally produced ceramic filter for Gunungkidul, a commercially available pot-shaped and silver-treated filter manufactured by *Pelita Indonesia* was analyzed. Based on this analysis and observations in field studies, possible drawbacks were identified and addressed in further studies for the development of an optimized locally produced filter for use in Gunungkidul.

**Morphology and composition.** The porosimetry showed that the pores were too big to retain bacteria by size exclusion. However, several mechanisms such as depth straining (Hutten, 2007), particle bridging (Cheremisinoff, 2002), inertial impaction, interception, diffusion and electrostatic attraction (Hutten, 2007) are important particle capture factors in depth filtration. The disinfection mechanisms of the silver nitrate coating also contribute to bacteria reduction.

With about 11 %, the ceramic contained a relatively high amount of  $\text{Fe}_2\text{O}_3$ , suggesting an iron-rich clay, which results in the filter's reddish color. Moreover, this iron oxide in combination with other fluxes may contribute to the formation of a liquid phase at low temperatures (Jasmund and Lagaly, 1993) and thus sufficient strength of the ceramic. The arsenic in the ceramic probably also came from the clay, although rice husks used as pore-forming agents might play a role. It is known that rice grown on arsenic-rich soil

and/or with contaminated water might also contain relatively high concentrations (Bhattacharya *et al.*, 2009). Bhattacharya *et al.* found arsenic concentrations in rice husks of up to 1.35 mg/kg dry weight (Bhattacharya *et al.*, 2009). Further analysis of the source materials would be necessary as a basis for recommendations for production of filters containing less arsenic.

**Flow rate and hydraulic conductivity.** The hydraulic conductivities ( $k$ ) of the *Pelita* filters in this study were between 1.5 and  $4.4 \times 10^{-7}$  m/s. This is higher than those observed by Van Halem (1.3 to  $1.37 \times 10^{-7}$  m/s) and Annan *et al.* ( $1.46 \times 10^{-7}$  m/s), but consistent with those described by Oyanedel-Craver and Smith, who observed hydraulic conductivities of 1.15 to  $5.01 \times 10^{-7}$  m/s (Van Halem, 2006; Oyanedel-Craver and Smith, 2008; Annan *et al.*, 2014).

*Pelita* aims to achieve a flow rate of 1.5 to 2.5 L/h (Pelita, 2014). The initial flow rate in the experiments complied with this, but the rate quickly dropped to about 1.3 L/h. Nevertheless, the filter produces well over 0.475 L/h, which is the minimum needed to produce sufficient drinking water assuming 24-hour usage, for an average family size (4 persons) and consumption of 2 liters/person/day (Lantagne, 2001). Even after one year of daily use in Indonesia, the flow rate (0.8 L/h) was still sufficient to supply a family of four.

**Fouling.** The long-term field study in Gunungkidul showed that after two years of use, the flow rate was down to 0.2 L/h. There was no biofilm visible, loss on ignition was about 1 % and incident light microscopy revealed a layer of salt crystals at the inner filter surface and additional crystals in the pores. Thus, the formation of crystals rather than an organic layer seems to have the main fouling effect. These salt crystals probably derive from the high calcium carbonate concentration in the source water.

**Physico-chemical performance.** The analysis for physico-chemical performance showed that pH, EC and hardness were consistent with guideline values. The same applied to nitrite ( $\text{NO}_2^-$ ), whose concentration never exceeded guideline values. The initial leaching of nitrate ( $\text{NO}_3^-$ ) was probably due to the silver nitrate coating, although it did not exceed the guideline value of 50 mg/L (WHO, 2011a). Arsenic, manganese and silver were leached from the filter. There are no guideline values for silver in the WHO drinking water regulations. However, silver concentrations of 0.05 mg/L or higher can be



observed in drinking water disinfected with silver (WHO, 2011a). German drinking water regulations allow a concentration of 0.08 mg/L for water that was disinfected with silver (DVGW, 2001). Thus, silver concentrations of 0.02 mg/L are acceptable in the filtrate. The health based recommended maximum value for manganese of 0.4 mg/L (WHO, 2011a) was never reached. The only hygienically critical value in the study was that of arsenic in the filtered water, which was above *WHO's* threshold of 0.01 mg/L (WHO, 2011a). However, the concentration of arsenic in the filtrate decreased after several liters of water had been filtered. In F7 (after 33.5 L), the value was down to 0.018 mg/L. The *WHO* guideline value – 0.01 mg/L – is provisional, however, and *WHO* state that “... every effort should be made to keep concentrations as low as reasonably possible” (WHO, 2011b). In summary, a filter containing less arsenic would be desirable.

Another possible approach is preventing arsenic leaching from the filter by higher firing temperature or using fluxes like illites or feldspar, or a combination of both, to bind the arsenic into the glass phase. Further investigations are needed, particularly for those cases where the concentrations of leached arsenic or other elements are high. However, in case of the filter analyzed in this study, an intensive flushing of the filter before use reduces the arsenic concentration in the filtered water effectively.

Analysis of filtrates of an old (2 years in use with about 13.300 L) and new filter (brand new, first 7 L) showed that the old filter was able to reduce calcite solubility significantly. However, as the samples taken in Indonesia could not be stabilized for transport, the results give an idea on the water properties but cannot be regarded as 100 % reliable. Thus, the reported reduction in EC might be influenced by adsorption mechanisms during transport, although both samples (raw water and filtrate) were transported to Germany in the same way. Increased reduction in EC and hardness by the old filter might be due to changes in the pore surface of the filter that led to an increased reduction of ions. This was also confirmed by calcite values which were about 98 mg/L in the raw water and 54 mg/L in the filtrate of the old filter (new filter 65 mg/L) (complete data in appendix).

**Microbial performance.** Log reductions for bacteria were about 3 to 5 log steps and thus consistent with values described in literature such as those observed with *E.coli* by Lantagne *et al.* (2010), which were about 3.1 to 6.1. For phages the log removal (0.3-

0.9 log steps) was comparable to values reported by Lantagne, which were about 0.09 to 0.5 log steps for filters produced in Nicaragua (Lantagne, 2001).

The WHO guidelines for evaluating household water treatment options describe a bacteria log reduction of 2 as protective and 4 as highly protective (WHO, 2011c). Thus, on average, the *Pelita* filter can be described as highly protective against bacteria. For viruses the guidelines describe a log reduction of 3 as protective and 5 as highly protective (WHO, 2011c). With a log reduction below 1, the filter exhibited very low removal efficiency for viruses. In literature, it is noted that metal oxides mixed with the clay before firing might lead to a better absorption capacity for viruses (Brown and Sobsey, 2009). One of the suggested metal oxides was  $\text{Fe}_2\text{O}_3$ , which is one of the main components of the filter tested. However, in this study insufficient virus removal was achieved. The effect of metal oxides in ceramic filters on virus removal needs to be addressed in further studies.

The results are published in Matthies *et al.* (2015).

In general, the ceramic filter was well accepted by the local people and field observations as well as laboratory studies showed that this sort of filter has the potential to reduce waterborne diseases as it is protective against hygienically relevant bacteria. Thus, ceramic filters proved to be an appropriate technology for Gunungkidul. However, *Pelita* filters are mostly promoted in another part of Java. Thus, a local production of ceramic filters is needed in order to assure accessibility for the local population and a sustainable use of the filters. A local production with locally available material is not only sustainable but also promotes the local industry. Moreover, results showed that care should be given to arsenic leaching, virus removal capacity and recontamination due to handling mistakes. Thus, if possible, arsenic-poor raw material should be used in order to minimize arsenic leaching, filters should be optimized for bacteria and virus removal and an optimized design should be tested in order to prevent recontamination due to handling mistakes. Fractioned filtration should be tested as an alternative to silver treatment of the filters and to minimize risk of recontamination.

## 4.2 Development of an optimized filter to be locally produced

Based on the drawbacks that were previously identified in the experiments with the *Pelita* filter, it was decided to test an optimization and establish a local production of the filter. To establish an optimized and sustainable ceramic filter for use in Gunungkidul, an appropriate composition and firing procedure needed to be developed. Therefore, the local raw material and firing habits as well as the performance of filters depending on changes in composition and design were analyzed. As previously described, particular attention was given to possible drawbacks identified in the analysis of an existing ceramic filter produced by *Pelita*.

### 4.2.1 Development of an appropriate composition under consideration of local raw material characteristics and performance

**Raw material.** The raw material typically used in the local clay mixtures showed a high concentration of iron oxide (about 13 % in the clay mixture) which resulted in a reddish color of the ceramic. This high concentration of iron and other mineral impurities is typical for many earthenware clays (Jasmund and Lagaly, 1993; Dies, 2003). As the clay mixture is adjusted according to the feeling of the potters and composes of varying clays from different sources, it is virtually impossible to give an exact composition for the clay mixture. All mixtures produced by the potters that were analyzed in this study seemed suitable as a base for preparing ceramic filters. However, in order to assure more continuity and thus uniform product quality, it is recommended to use large clay deposits that do not have to be changed repeatedly.

**Physico-chemical performance/ leaching.** In order to determine significant impacts of the filters on water quality, physico-chemical analyses of raw water and filtered water were conducted. During the experiments, ceramic filtration led to a slight increase in pH but no significant changes in EC or hardness. However, after a few batches were filtered, calcite solubility in the filtrates decreased which was similar to the results obtained from the long-term field experiment in Gunungkidul. Here, it was visible that lime precipitated in the plastic receptacles of the filtration unit and less lime deposited in the cooking pots. This could be due to the decreased calcite solubility resulting from changes in the pore surface of the filter that led to changes in ion concentration. This

side-effect would be very desirable as the main concern of many local people is the hardness of the tap water. A decrease in hardness would contribute to the acceptance of ceramic filters.

Experiments with natural brook and river water showed that filters effectively reduced turbidity and TOC. Similar to the *Pelita* filter, arsenic and manganese were leached from the ceramic filter pots. Initial concentrations were varying from 0.001 to 0.036 mg/L for arsenic and <0.01 to 0.72 mg/L for manganese. Thus, values partly exceeded the *WHO* guideline threshold value of 0.01 mg/L for arsenic and the *WHO* health based value of 0.4 mg/L for manganese (WHO, 2011a). However, concentrations decreased after filtration of several liters so that leaching of arsenic and manganese did not pose an actual health risk. Nevertheless, a thorough flushing of the filters before use is advised to prevent uptake of increased concentrations. Moreover, in the first filtrates, an increased turbidity, color absorption, EC, calcite solubility and concentration of TOC, sodium, potassium, boric, nitrite, ammonia and sulfate could be observed. In discarding the first two - three batches, people will avoid taking up those substances that are washed from the filter. Arsenic, which was leached most slowly, was below the *WHO* guideline value after filtration of 23 batches (here about 26 L).

**Porosity.** The fired ceramic without addition of pore-forming agents showed a porosity of about 36 % with a median pore size of 0.03  $\mu\text{m}$  which was much smaller compared to the data obtained from the *Pelita* filter with 8.34  $\mu\text{m}$  pore size and about 49 % porosity. As possible pore-forming agents, sawdust and rice husks were compared as they are best available in the project region.

Sawdust generally resulted in higher porosities and median pore sizes compared to the same concentration of rice husks. However, the use of rice husks led to formation of some pores with higher diameters. In general, the pore size distribution of the ceramics made of sawdust or rice husks was very similar to the distribution obtained from the *Pelita* ceramic filter. Comparing the porosity and median pore sizes of the filters with the *Pelita* filter, with a median pore size of 9.93 and 47 % porosity the filter containing 40 % rice husks was closest to the data obtained from the *Pelita* filter. However, the surface of the filters made of rice husks was not as smooth as sawdust filters and at 40 % pore-forming agents, filters with rice husks were more brittle compared to those made of the

same concentration of sawdust. Thus, though both pore-forming agents seemed suitable as combustible material, rice husks should be used at less than 40 %.

An additional screening of the material could help preventing extreme high pore sizes though it requires additional tools, effort and monitoring. However, to assure a uniform filter quality, a thorough screening of the combustible material is inevitable. It is known that apart from too large pore-forming particles causing too high porosity and fragility, too small particles lead to chalky filters. Therefore, screening with two sieves and using the middle fraction is recommended (Dies, 2003).

Moreover, it should be noted that pore-forming agents should be added to the clay mixture (different clays, sand, water) after the mixture has been finished because otherwise the pore-forming agents will absorb too much of the water (Dies, 2003).

**Flow rate.** Flow rates of unused filters with 40 % pore-forming agents showed no big differences between sawdust and rice husk filters. To enable comparability with ceramic filters of different capacities, the hydraulic conductivity was calculated. For the unused filters made of 40 % combustibles, the conductivity was about  $1.5$  to  $2 \cdot 10^{-6}$  m/s. Used filters had hydraulic conductivities of about  $1.1 \cdot 10^{-6}$  for the rice husk filter and  $2.6 \cdot 10^{-7}$  m/s for the sawdust filter. However, the rice husk filter was used for 50 batches, while the sawdust filter was used for 83 batches when the measurements were done. Thus, the used 40 % filters were not directly comparable. Used 20 % filters (about 40 to 50 batches) showed lower hydraulic conductivities of  $1.9$  to  $2.1 \cdot 10^{-7}$  m/s without significant differences between rice husks and sawdust. The filter without pore-forming agents had a hydraulic conductivity of  $6.8 \cdot 10^{-8}$  m/s which was lower compared to filters with combustibles. In literature, hydraulic conductivities of *PPF* filters were about  $1$  to  $5 \cdot 10^{-7}$  m/s (Van Halem, 2006; Oyanedel-Craver and Smith, 2008; Annan *et al.*, 2014), and the filter obtained from *Pelita* showed conductivities of  $1.5$  and  $4.4 \cdot 10^{-7}$  m/s (Matthies *et al.*, 2015). Thus, the flow rate of the filter without combustibles was too low. However, a filter made of 10 % pore-forming agents could be tested as it might still have sufficient flow rate and show an increased log reduction compared to the filters made of 20 % combustibles. Assuming a design similar to the *Pelita* filter (about 8 L capacity), a hydraulic conductivity of  $1 \cdot 10^{-7}$  m/s is necessary to achieve a flow rate of 0.475 L/h. Assuming 24-hour usage, this is the minimum flow rate needed to produce sufficient drinking water for an average family size (4 persons) and consumption of

2 liters/person/day (Lantagne, 2001). Thus, the hydraulic conductivity and flow rate of filters made of 20 % combustibles were best in range with reported values and would produce enough drinking water for an average family size.

**Correlation of pore-forming agent concentration and microbial log reduction.**

Relation of pore-forming agent concentration and removal efficiency for bacteria and phages was determined with sawdust filters. Heterotrophic plate counts in the brook water experiments were increased in the filtrates. No significant differences could be detected in filters with different pore-forming agents. However, coliform bacteria and enterococci present in the brook decreased during filtration. Population analysis showed that all filter types exhibited distinct population patterns, indicating different removal capacities on bacterial groups. It needs to be remarked that changes in the band pattern not necessarily mean that the presence of bacteria changed. It is also most likely that the pattern changed when a certain species was removed as the DNA of certain species is better amplified when other species are eliminated. Sequencing results revealed that detected bacteria were harmless environmental bacteria belonging to the beta and gamma proteobacteria groups. No dependency of the band pattern on shape or size of the bacteria could be detected.

To further analyze the removal capacity on hygienically relevant bacteria, experiments were conducted with water spiked with *E. coli*, *Ent. faecium* and *P. aeruginosa*. As expected, bacteria log removal increased with decreasing concentration of combustibles as lower porosity and lower pore sizes impeded the passage of bacteria. For *Ent. faecium*, removal was 0.5 to 0.6 log steps for the filter made of 40 %, 1.3 to 1.8 log steps for the filter made of 20 % and 1.9 to 2.4 log steps for the filter made of 0 % combustibles. With a log reduction of 2, only filtration through the filter without combustibles could be described as protective against potentially pathogenic bacteria (WHO, 2011c).

The increasing heterotrophic plate counts and decreasing counts for health-relevant bacteria at the same time, suggest that some bacteria are comfortable in the ceramic structure. The removal of several species probably also contributes to a growth of remaining bacteria.

The typical size of bacteria and non-viral pathogens is between 1 to 3  $\mu\text{m}$ , while viruses have a size of about 10 to 100 nm (Willey *et al.*, 2008). With many pores of a diameter

between 10 to 100  $\mu\text{m}$ , the pores of the filters were too big to retain bacteria and viruses simply by size exclusion which was also the fact for the *Pelita* filter. Thus, mechanisms such as depth straining (Hutten, 2007), particle bridging (Cheremisinoff, 2002), inertial impaction, interception, diffusion and electrostatic attraction (Hutten, 2007) are important factors in bacteria and virus removal during filtration.

For phages, log removal was highest when using filters with 40 % sawdust (0.4 to 0.6 log steps). The filter made of 20 % sawdust showed almost no reduction (0 to 0.1 log steps) and the filter without combustibles showed a 0.1 to 0.3 log reduction. Results suggested, that adsorption of phages within the ceramic structures was higher at higher porosities.

The amphoteric nature of phages leads to an adsorption at charged surfaces. As metal oxides are negatively charged, they contribute to an adsorption of phages at the ceramic surface (Brown and Sobsey, 2009). In case of the analyzed filters, iron oxide in form of hematite and maghemite was present in the clay in high concentration which suggested an improved removal capacity for phages. However, in the ceramic fired at 850  $^{\circ}\text{C}$ , no hematite and maghemite could be detected due to mineral phase transition during firing. Thus, further research is needed on the relation of phage log reduction as well as nature and concentration of metal oxides in the clay and resulting ceramic.

In general, none of the tested filter types met the *WHO* recommendations for virus reduction. For bacteria removal, only the 0 % filter proved to be protective against bacteria. However, the low flow rate of this filter was too low for POU water treatment. Therefore, fractioned filtration and silver coating were tested as approaches to further improve bacteria and virus reduction.

#### **4.2.2 Development of an appropriate firing procedure under consideration of local firing habits**

Examination of local firing habits showed that, traditionally, the earthenware clays in the local pottery village Kasongan are fired at temperatures of about 600 to 750  $^{\circ}\text{C}$ . However, temperatures vary between batches because potters generally add wood to the fire according to their feeling and adjust it according to the size of the pottery.

Moreover, temperatures vary within the kilns as they have an open ceiling which is covered with any available material and most of them have cracks resulting from an

earthquake in 2006. Thus, actual kilns cannot keep the heat properly and are very inefficient.

The heating rate observed in this study varied between 7 and 13 K/min. This very fast heating was possible due to the relatively small amount of quartz in the clay mixture. At 573 °C, the so-called quartz inversion leads to a high increase in volume which is reversible (Salmang and Scholze, 2007). Therefore, this temperature should be passed slowly at about 5 K/min to prevent formation of cracks. The cooling rates were quite fast as well, but tolerable.

According to literature, typical firing temperatures for ceramic filters range from 550 (Archer *et al.*, 2011) to 832 (Pelita, 2012), 870 (Lantagne *et al.*, 2010) and 900 °C (Oyanedel-Craver and Smith, 2008; Van Halem, 2006) or even higher. Melting analysis of the clay mixture showed that sintering started at a temperature of 864 °C. However, comparative analysis of the ceramics produced in the gradient firing kiln at temperatures from 550 to 1050 °C and of pots fired locally at about 650 and 800 °C showed that the ceramic was already very strong when fired at low temperature. This could be due to the high proportion of iron hydroxide, iron oxides and other mineral impurities in the raw material that form a liquid phase at lower temperatures (Verkerk *et al.*, 1982; Jasmund and Lagaly, 1993; Dies, 2003). Shrinking, bulk density and water absorption were stable up to a firing temperature of 750 °C, suggesting that firing at higher temperatures leads to higher density and thus lower flow rates. Thus, a compromise between strength and flow rate needs to be made.

Generally the local firing procedures can be used for production of ceramic filters. However, as temperatures throughout the kilns showed strong variations due to the open design and cracks in the walls, adjustments are recommended. In order to assure a homogenous firing temperature, and thus filter quality, a more advanced construction with sufficient insulation would be desirable. One such advanced construction available in Kasongan is shown in the appendix (Figure A 8). It has a fixed roof and a chimney. This kiln was built in a charity project after the earthquake in 2006, but has never really been in use. Recently, the kiln was damaged in a storm but it could be restored and used for filter production. However, people will need training in proper use of the kiln.



Such an advanced kiln, if operated appropriately, would also use less wood to achieve the same or higher firing temperature as it works much more efficient compared to the open designs that are more popular in Kasongan.

Another possibility to achieve reproducible quality of ceramic filters would be the use of already existing gas-fired kilns. However, a cost-benefit evaluation needs to be done.

Moreover, a temperature control is required to control the firing temperature and thus attain a constant quality of ceramic filters.

#### **4.2.3 Development of an optimized design under consideration of filter performance**

**Fractioned filtration.** Bacteria log reduction for **rice husk filters** increased with subsequent filtration steps from 0.5 (40 %) to 1.2-1.5 (40+20 %) and to >3 log steps (40+20+0 %) for *Ent. faecium*. Fractioned filtration (40+20+0 %) achieved about 1.7 to 1.9 log steps for *E. coli* to 2.4 to >4.5 log steps for *P. aeruginosa*. Thus, for rice husk filters, a fractioned filtration proved to be quite efficient and with log reductions above 2.4 to 4.5 protective to highly protective against *Ent. faecium* and *P. aeruginosa* (WHO, 2011c). For *E. coli*, the log reduction was just below 2 log steps.

For **sawdust filters**, fractioned filtration log reductions were generally lower compared to rice husk filters which could be due to the increased median pore sizes and porosities in sawdust filters. Moreover, the third filtration step through the filter without combustibles did not lead to an increased log reduction compared to two-step filtration (1.3 to 1.7 log steps for *Ent. faecium*) although single filtration through this filter showed the highest log reduction for sawdust filter experiments (1.8 to 2.5 log steps for *E. coli*, 2.6 to 3.2 log steps for *P. aeruginosa*). Respectively, two-step filtration did not show a benefit compared to single filtration through the filter made of 20 % sawdust. Thus, for sawdust filters, fractioned filtration showed no benefit compared to single filtration through the filters made of 20 or 0 % sawdust.

For **phages** which are used as surrogates for viruses, log reduction was highest for fractioned filtration (40+20+0 %) with 0.8 to 1.0 log step for rice husk filters and 0.5 to 0.9 log steps for sawdust filters. Here again, rice husk filters showed slightly higher removal capacities. However, with log reductions up to 1 log step, filters were not protective against phages.

Taking into account that the filter without combustibles is not feasible as its flow rate is too low, filters made of 20 % pore-forming agents seem to be the most efficient system for local conditions. Both rice husks and saw dust seem suitable as pore-forming agents. A fractionated filtration through a filter made of 40 % before this coated filter made of 20 % combustibles would give an additional benefit for virus removal and would protect the second filter from turbid particles. Thus, this main filter is additionally protected from fouling and needs to be cleaned less often which reduces the recontamination due to handling mistakes during cleaning.

**Silver coating.** Usually, ceramic filters based on the *PFP* design are treated with silver in order to increase the bacteria removal capacity. In general, silver can be applied to the filters by coating silver solution onto the surface or by submerging the filters into a silver solution. It is reported that the application method of silver is less significant than the applied silver concentration. However, Oyanedel-Craver and Smith observed that bacteria removal was higher when the filter was dipped into the silver solution compared to coated filters (Oyanedel-Craver and Smith, 2008). For submerging the filters, a bigger volume of silver solution might be needed whereas an even coating is more complicated to achieve. Recommended application forms are coating with 6 mL of a 200 g/L colloidal silver solution for a filter diameter of 6.5 cm or submerging the filter in a 800 mg/L colloidal silver solution for 45 s (Oyanedel-Craver and Smith, 2008).

Submerging the filters into a silver solution seems to be more reproducible than coating the surface with the silver solution as the surface soaks the fluid very fast and a homogenous coating is difficult to attain. Therefore, submerging of the filters in a silver nitrate solution was chosen as application method.

Filters made of 20 % sawdust with silver treatment achieved log reductions of above 4 to 5 log steps for bacteria and about 1.2 to 1.9 log steps for phages which were both at least 1 log step higher compared to non-coated filters. Thus, silver had a significant effect on bacteria and phage removal. The disinfection effect of silver against bacteria is mainly based on the interaction of the silver with nucleic acids, the silver-induced changes in the cell membrane structure and the reaction of silver with thiol groups in functional proteins and structural groups of bacteria (Russell and Hugo, 1994). Respectively, silver ions react with viral DNA and thiol groups in functional proteins and

thus inactivate the virus (Kim *et al.*, 2008). However, generally, viruses are not inactivated at low silver concentrations typically used in POU water treatment and bacteria may develop resistance mechanisms against silver (Sobsey, 2002). Thus, inactivation probably does not result from silver alone, but from an interrelation of several mechanisms. Nevertheless, silver-coated filters were highly protective against bacteria but not protective against viruses (WHO, 2011c).

Another aspect supporting the use of silver is the varying composition of the local clay mixture and corresponding filter quality. The silver treatment poses an additional disinfection effect, so that sufficient bacteria reduction can be secured despite of varying ceramic quality.

It has to be mentioned however, that the inactivation mechanism of silver is not yet fully understood and some bacteria are known to fall into a non-culturable but metabolic active state in the presence of silver (Königs *et al.*, 2015). Thus, further experiments should be done in order to confirm that bacteria are actually killed.

Moreover, the use of silver as disinfectant is controversial as it might have toxic effects on the human health when the body is continuously and extensively exposed to silver ions (Fung and Brown, 1996; Gulbranson *et al.*, 2000; Drake and Hazelwood, 2005; Panyala *et al.*, 2008). Some researchers even describe the toxic effect of silver on bacteria to be comparable to its toxicity in human cells (Greulich *et al.*, 2012). Thus, handling highly concentrated silver solutions and consuming water with enhanced silver concentrations might be harmful to manufacturers and users. Therefore, leached silver concentrations in the filtrates were analyzed.

Initially, leached silver concentration from the silver-treated filter was about 0.014 to 0.024 mg/L which decreased to <0.003 mg/L after 22 L were filtered. For water that was disinfected with silver, the German drinking water regulations allow a concentration of 0.08 mg/L (DVGW, 2001). There are no guideline values for silver in the *WHO* drinking water regulations. However, silver concentrations of 0.05 mg/L or higher can be observed in drinking water disinfected with silver (WHO, 2011a). The *WHO* further estimates that silver levels in drinking water can be tolerated up to a concentration of 0.1 mg/L. As a base for this value, they take the NOAL (no-observed-adverse-effect level) which is 10 g over a life time of 70 years (WHO, 2003). Thus, even initial silver concentrations were acceptable. Though silver leaching does not pose a health risk for

the users of the filters, the health of the manufacturers that handle silver solutions with high concentrations for coating the filters is threatened. It has to be ensured that safety measures are kept.

Apart from fractioned filtration (see previous paragraph) as an alternative to silver treatment of the filters, the use of metal oxides to increase removal capacity for viruses might be interesting. One possible oxide described in literature is  $\text{Fe}_2\text{O}_3$  which is mixed with the clay before firing. These metal oxides might lead to an increased absorption capacity for viruses (Brown and Sobsey, 2009). Another possible approach would be the use of antibacterial nano particles or allophanic material in combination with the ceramic filter. However, most existing approaches also use it in combination with silver (Onodera *et al.*, 2001; Quang *et al.*, 2011; Quang *et al.*, 2013). Moreover, the approaches using nano particles are quite expensive and handling is complicated and not feasible in Gunungkidul. However, further research on these alternatives was not possible in the frame of this dissertation.

**Fouling.** In experiments with very turbid brook water (28 NTU), a layer of particles formed at the bottom of the filters made of 40 % rice husks. Fractioned filtration showed that the subsequent filters were mostly not affected by turbid particles from the brook water. The first filter was able to remove most of the suspended particles, so that the turbidity was down to about 0.23 NTU. The flow rate of this filter made of 40 % rice husks decreased from 1.5 L/h to 0.5 L/h due to the cake layer formed at the bottom and particles clogging the pores. However, this layer was mostly formed at the bottom of the filter as particles settled during filtration. Moreover, flushing with clean tap water and brushing with a soft brush was very efficient and flow rate could be restored. Flow rate of the filters made of 20 % and 0 % rice husks was also increased after cleaning though it had not been decreased through filtration of brook water. As the filters had been used before (about 23 batches), particles had probably already induced a decrease in flow rate and were removed through cleaning. Thus, cleaning with unpolluted water and a soft brush proved to be very efficient to restore flow rate. However, long-term field studies with the *Pelita* filter showed that after two years of use, the flow rate was not sufficient anymore. Refiring at 850 to 900 °C would be a possibility to restore the flow rate, as not only organic material but also calcium carbonate is burned at this temperature.

**Optimized design.** Taking into account the flow rates and log reductions, filters made of 20 % pore-forming agents that are treated with silver nitrate seem to be most efficient. Filters without combustibles had a relatively high bacteria removal but flow rate was not sufficient to supply a family of four. Fractioned filtration with a pre-filtration step through a filter made of 40 % combustibles prior to this coated filter made of 20 % combustibles would give a benefit for virus removal and would protect the coated filter from turbid particles and thus a fouling of this main filter.

This filter made of 40 % combustibles should be cleaned regularly by the users with purified water (produced by the filter) and a soft brush to remove a potential particle or biofilm layer clogging the pores. Thus, the main filter does not need to be cleaned regularly and the clean side of this filter is protected from recontamination during cleaning. Once the flow rate of the pre-filter is not restored any more by cleaning, the pre-filter can be exchanged. Should the main-filter be blocked through organic material, it could be brought to the pottery for refiring and used again. However, in case of the filter analyzed in a long-term field study in this work, the clogging was mainly due to inorganic material. However, if calcium carbonate crystals were the main reason for clogging, refiring might still be efficient in restoring the flow rate as calcium carbonate is burned as well. Though recycling is less expensive compared to buying a replacement filter element, investment costs will be higher if the filter composes two elements. Thus, in addition to long-term field studies, a thorough economical analysis needs to be made for this fractioned design option.

Furthermore, once the reservoir of a household will be equipped with a gravel filter at an outlet tap as planned in the master plan, further pre-filtration for removal of turbidity might probably not be needed. Thus, the water can be directly poured into the silver-coated ceramic filter made of 20 % combustibles.

**Handling, acceptance and price.** A field study in Gunungkidul with commercial filters produced by *Pelita* revealed that the filter was generally well accepted. However, an intensive training of the users was needed to assure a correct handling and prevent recontamination. Apart from an optimized design which contributes to a safe handling, other issues should be addressed.

To assure a wide usage of filters, several obstacles need to be overcome. It is of utmost importance to raise awareness for contamination of water, hygiene and contamination

pathways. Otherwise, many people don't see the need to act. Apart from easy handling, the filter should be affordable and easily available for everyone.

According to literature, *PPF* filters globally cost about US\$ 5-25. For instance, the filter produced in Cambodia was about US\$ 10 for the complete filter unit (replacement filters US\$ 2.5-5) in 2007 (Brown, 2007). In 2012, filter units produced by *Pelita* cost about IDR 125,000 to 250,000 (replacement filters IDR 85,000) which was equivalent to about US\$ 13.7 to 27.3 for the whole filter unit (replacement filters US\$ 9.3). Interviews with local people, especially the secretary of the pilot village Pucanganom (Pak Suprpto, Sekretaris Desa Pucanganom) and the former chief of the Gunungkidul branch of the *Indonesian Red Cross* (Drs. Sumedi, Ketua *Palang Merah Indonesia* Cabang Gunungkidul) suggested that normal poor farmers in the region would not spend more than IDR 50,000 (US\$ 5.5) for the replacement filter element. On the other hand, some people drive long distances to buy bottled water for about IDR 12,000/19 L (US\$ 1.3). For them, the ceramic filter already is an efficient and cheap alternative. Though people will be ready to pay more when awareness for water contaminants and hygienic contexts is raised, in order to guarantee a wide usage of the filters in Gunungkidul, prices should be kept as low as possible.

Within this work, contact was made with the local Gunungkidul branch of the *Indonesian Red Cross* as well as the NGO *Yayasan Dian Desa* in Jogjakarta. Both organizations were very interested in the work on locally produced ceramic filters and could be very helpful in establishing a distribution of the filters.

**Transferability.** With about 685,210 inhabitants (Badan Pusat Statistik, 2008) without access to safe drinking water, the Gunungkidul district poses a big potential sales opportunity for the ceramic filter as an appropriate POU water treatment option. However, the filter could also be distributed to other regions in Java and whole Indonesia where people live in a similar situation. The approach used in this work to develop a composition for local production of ceramic filters can be transferred to all regions where safe drinking water is not available and POU water treatment poses a great opportunity to produce safe drinking water for the local population.

## 5 Summary and conclusion

POU water treatment is the only feasible way to get safe drinking water for most people in Gunungkidul. Ceramic pot filters based on the *PFP* technology seem to be most appropriate in the project region. The aim of this work was the development of an appropriate composition and firing procedure for the local production of ceramic filters for household water treatment. The analysis of a commercially available ceramic filter revealed that ceramic filters are generally well accepted by local people, but intensive training is needed to assure a correct handling of the filters. Further drawbacks revealed during the analysis were leaching of arsenic and manganese as well as virus removal capacity.

To develop an appropriate composition and firing procedure, locally available raw material and local firing habits were analyzed. Local clays were generally suitable for the production of ceramic filters. They mainly comprised silica, alumina and iron oxide in form of mineral phases hematite, maghemite, calcite and a huge group of quartz and silicates.

Traditional firing habits at temperatures of about 600 to 750 °C proved to be sufficient for production of ceramic filter pots. Filters were quite stable even at low firing temperatures. Most probably, iron oxide and other impurities in the local earthenware clays allow a firing at low temperatures. However, the use of more advanced firing kilns with better insulation and heat efficiency would be desirable. Such kilns are also available locally, but might need some adjustment. However, potters need to be trained in operation of those kilns. Moreover, the use of temperature controls is suggested in order to assure a consistent ceramic quality.

As combustible materials, sawdust and rice husks proved to be applicable. However, a screening using two sieves is desirable to achieve a more homogenous pore size distribution. Porosity of ceramics without pore-forming agents was about 36 % with a median pore diameter of 0.03 µm. A filter made of this clay mixture had a hydraulic conductivity of about  $6.8 \cdot 10^{-8}$  m/s which, assuming a design similar to the *Pelita* filter, is too low to produce sufficient drinking water for a family of four. With 40 % sawdust or rice husks, porosity was increased to about 50 % with a median pore size of about 10 µm and a resulting hydraulic conductivity of about  $2.6 \cdot 10^{-7}$  to  $1.1 \cdot 10^{-6}$  m/s. 20 %

pore-forming agents resulted in ceramic filters with median pore diameters of 1.8 to 3.5  $\mu\text{m}$ , a porosity of about 44 % and a hydraulic flow rate of 1.9 to  $2.1 \cdot 10^{-7}$  m/s. This was best in range with hydraulic flow rates reported from other *PPF* filters.

Though heterotrophic plate counts were increased in the filtrates, hygienically relevant bacteria were reduced. For bacteria removal studies, relevant potentially pathogenic bacteria (*E. coli*, *Ent. faecium* and *P. aeruginosa*) were chosen based on the *WHO* guidelines for *Evaluating household water treatment options* (WHO, 2011c).

As expected, bacteria removal efficiency decreased with increasing porosity. Fractioned filtration did not necessarily show a benefit in bacteria removal compared to single filtration through the respective ceramic filters with lowest porosity. In contrast, virus removal capacity was highest with the filters made of 40 % combustibles and fractioned filtration showed an increase in log removal capacity. Silver coating proved to increase the bacteria and virus removal capacity significantly.

In conclusion, taking into account the flow rates and log reductions, filters made of 20 % pore-forming agents that are coated with silver seem to be the most efficient system for local conditions. A fractioned filtration through a filter made of 40 % combustibles before this coated filter made of 20 % combustibles would give a benefit for virus removal and would protect the second filter from turbid particles, and thus, a fouling of this main filter. The pre-filter should be cleaned regularly with purified water and a soft brush. Thus, the main filter does not necessarily need regular cleaning and the clean side of this filter is protected from recontamination during cleaning. The combination of pre-filtration through a filter made of 40 % sawdust with subsequent filtration through a silver nitrate-coated 20 % sawdust filter showed a removal capacity of more than 4 to 5 log steps for examined bacteria and 1.6 to 1.9 log steps for viruses (surrogate: phage  $\phi\text{X174}$ ).

Summarizing, this work gives a thorough approach for the aspects that need to be addressed when establishing a new ceramic filter system. Available raw material for clay mixture and pore-forming agents need to be analyzed and substances that are leached from the ceramic filters need to be identified. The composition needs to be optimized concerning flow rate and removal efficiency for bacteria. If the source water is heavily contaminated with viruses, filters need to be optimized for virus removal. Depending on source water quality, special focus should be given to physico-chemical performance, for instance removal capacity of TOC or pollutions such as pesticides, where ceramic



filtration might not be an appropriate technique. Last but not least, ceramic filters need to be accepted by local users and awareness for water contaminants and hygienic contexts needs to be raised. Moreover, users need intensive training in handling the filter. Thus, capacity development is one of the most important aspects when establishing a POU treatment option such as the ceramic water filter.

## 6 Outlook

This work gives a thorough base for the implementation of a locally produced ceramic filter for Gunungkidul. However, there is still some need for research on further refining (sieves) and testing other concentrations of combustibles (e.g. 10 or 30 %) and long-term studies of the optimized, locally produced filters for bacteria and virus removal as well as fouling. To prevent leaching of arsenic and manganese, binding the elements into the glass phase by firing at higher temperatures or using fluxes like illites or feldspar, or a combination of both, could be promising approaches. These need to be investigated in further studies.

Another aspect that needs to be clarified for the development of ceramic filters in further regions is the relation between arsenic concentration in clay sources and arsenic leached from the filters. Thus, a recommendation could be given for maximum tolerable arsenic concentration in clay sources.

Moreover, colloidal silver needs to be investigated as an alternative to silver nitrate and their availability in Jogjakarta needs to be verified. The effect of cleaning of the filters on silver leaching and bacteria removal should be further investigated.

As this work showed that a fractioned filtration through a filter with higher porosity followed by a silver-coated filter with lower porosity is desirable, a possible design should be tested for suitability and efficiency. Further studies need to focus on refiring effects for restoring flow rates of clogged filters.

Moreover, alternatives to silver coating should be tested as silver toxicity cannot be excluded.

The local Gunungkidul branch of the *Indonesian Red Cross* has been very interested in the research results and showed interest in a local distribution of the ceramic filters. Once the investigations are completed and the filter is produced, this organization as well as the NGO *Yayasan Dian Desa* in Jogjakarta could be very helpful in distributing the ceramic filters.

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## 8 Appendix

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**Figure A 1:** Preparation of ceramic filters at *Pelita*, Bandung.

Left: press apparatus. Right: filters drying in shelves. Pictures were taken in March 2012.

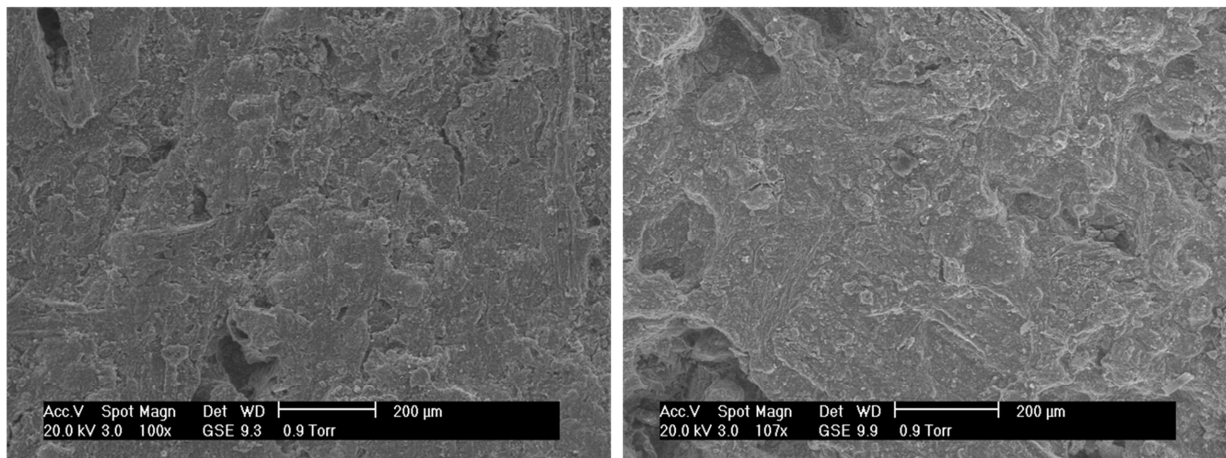


**Figure A 2:** Kiln at *Pelita*, Bandung.

Left: inside the old kiln. Middle: old kiln. Right: new kiln under construction. Pictures were taken in March 2012.





**Figure A 3:** ESEM pictures of outer (left) and inner (right) surface of *Pelita* filter.**Table A 1:** complete data for physico-chemical analysis of Pelita raw water and filtrates (as in Table 7)

Analyte concentrations for river water RW in Germany and field study in Indonesia. (RF = river water filtrate, IW = Indonesian raw water, IF old = filtrate from old filter, IF new = first filtrate from new filter.)

parameter	Germany		Indonesia		
	RW	RF	IW	IF old	IF new
pH [-]	7.66	7.92	7.4	7.95	7.54
EC [ $\mu$ S/cm]	470	457	504	270	519
Turbidity [NTU]	2.30	0.14	1.2	0.52	8.79
Boron [mg/L]	0.02	0.01	0.05	0.03	0.15
Cyanide [mg/L]	<0.005	<0.005	<0.005	<0.005	<0.005
Fluoride [mg/L]	0.12	<0.1	<0.1	<0.1	0.54
Chloride [mg/L]	15.9	17.3	3.4	3.8	12
Sulfate [mg/L]	24.1	27.3	1.9	1.9	35.7
Ammonium [mg/L]	<0.01	0.01	0.02	<0.01	7.2
Nitrate NO <sub>3</sub> <sup>-</sup> [mg/L]	7.71	7.80	8.3	9.7	21
Nitrite NO <sub>2</sub> <sup>-</sup> [mg/L]	0.08	0.09	<0.01	<0.01	25.5
Ortho phosphate [mg/L]	0.07	1.09	<0.03	<0.03	1.3
Total hardness [°dH]	9.7	10.2	15.9	8.8	11.8
Carbonate hardness [°dH]	8.0	8.0	14.8	8.6	8.6
Hardness [mg/L CaCO <sub>3</sub> ]	1.57	1.62	2.83	1.55	2.1
Acid capacity [mmol/L]	2.85	2.87	5.3	3.06	3.08
Oxidizability [mg/L]			1.13	0.73	14.7

Sodium [mg/L]	9.42	9.18	3.31	3.67	5.09
Potassium [mg/L]	2.0	2.19	1.28	1.49	7.17
Magnesium [mg/L]	7.6	8.19	14.1	4.87	11.7
Calcium [mg/L]	57.4	59.4	98.3	54.2	64.8
Calcite solubility [mg/L]	-18.0	-13.0	-31.41	-12.26	-4.33
Aluminum [mg/L]	<0.005	<0.005	<0.005	0.014	0.007
Antimony [mg/L]	<0.0001	<0.0001	<0.0001	<0.0001	<0.001
Arsenic [mg/L]	<0.001	0.045	<0.001	0.001	0.08
Lead [mg/L]	<0.001	0.002	0.004	0.002	0.005
Cadmium [mg/L]	<0.0003	<0.0003	0.0001	<0.0001	0.001
Chromium [mg/L]	<0.001	<0.001	<0.001	<0.001	<0.001
Iron [mg/L]	<0.01	<0.01	<0.01	<0.01	<0.01
Copper [mg/L]	<0.01	<0.01	<0.001	<0.001	0.014
Manganese [mg/L]	0.003	0.001	<0.01	<0.01	0.27
Nickel [mg/L]	<0.01	<0.01	<0.001	<0.001	<0.001
Selenium [mg/L]	0.001	0.001	<0.001	<0.001	0.001
Mercury [mg/L]	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
TOC [mg/L]	1.99	2.15	0.72	0.22	23
Color (absorption at 436 nm) [1/m]	0.35	0.27	0.06	0.07	2.3
Color (absorption at 254 nm) [1/m]	5.34	5.33	0.57	0.52	47.6

**Figure A 4:** Cooking pots and tea.  
Left: raw water. Right: filtered water.



**Figure A 5:** Handling of the filter.  
Left and middle: cleaning of filter and receptacle. Right: applying water to be filtered.



**Table A 2:** Chemical composition of local raw material  
Elementary oxides of samples soil, red clay 1 and 2, yellow clay 1 and 2, brown clay, lava sand and clay mixture. (LOI=Loss on ignition)

[% w/w]	Soil	Red 1	Red 2	Yellow 1	Yellow 2	Brown	Sand	Mix
SiO <sub>2</sub>	64.5	50.5	47.6	59.4	45.5	51.4	53.3	51.5
Al <sub>2</sub> O <sub>3</sub>	21.1	30.3	33.4	22.3	34.7	28.8	19.8	26.2
TiO <sub>2</sub>	0.81	1.22	1.45	0.88	1.45	1.42	0.92	1.1
Fe <sub>2</sub> O <sub>3</sub>	8.5	14.6	16.2	8.08	17.1	14.6	9.51	13.0
CaO	2.41	0.89	0.31	4.18	0.35	1.26	8.18	3.88
K <sub>2</sub> O	0.51	0.38	0.06	1.92	0.10	0.56	1.66	0.84
MgO	0.84	1.75	0.55	2.36	0.34	0.99	2.83	1.3
MnO	0.16	0.08	0.14	0.09	0.18	0.22	0.19	0.44
Na <sub>2</sub> O	1.06	0.07	0.04	0.52	0.06	0.50	3.17	1.47
Cr <sub>2</sub> O <sub>3</sub>	<0.01	<0.01	0.01	0.02	0.02	0.01	<0.01	0.01
P <sub>2</sub> O <sub>5</sub>	0.05	0.08	0.05	0.12	0.08	0.16	0.26	0.16
SO <sub>3</sub>	<0.01	0.03	<0.01	0.05	<0.01	<0.01	0.03	0.04
SrO	0.02	<0.01	-	0.02	-	-	0.06	0.03
V <sub>2</sub> O <sub>5</sub>	0.02	0.05	0.06	0.02	0.08	0.06	0.04	0.05
ZnO	0.01	0.01	0.01	0.02	<0.01	<0.01	0.01	0.01
ZrO <sub>2</sub>	0.02	0.02	0.03	0.03	0.03	0.03	0.02	0.02
LOI	10.6	14.8	14.0	12.0	14.7	13.5	2.26	9.27

**Table A 3:** Chemical composition of local raw material  
Aqua regia digestion with subsequent ICP OES analysis of eluate:

[mg/kg]	Soil	Red 1	Yellow 1	Sand	Sawdust	Rice husks
Al	79876	107450	66700	39800	805	42
As	<4	5	8	<4	<3	<5
Ba	258	135	76	63	65	<25
Ca	8494	4430	27100	2400	28980	720
Cd	<4	<4	<4	<4	<3	<25
Co	15	16	12	14	<3	<25
Cr	7	17	45	6	<3	<25
Cu	29	74	15	23	19	<25
Fe	51625	78400	44700	42500	839	66
K	1085	1880	6645	749	6263	3210
Li	5	24	88	5	12	<25
Mg	3599	7505	10775	2195	10257	1100
Mn	1178	580	625	547	30	65
Na	1091	436	2150	3710	1158	49
Ni	<4	9	20	<4	<3	<25
Pb	12	15	17	6	<3	<25
Sr	92	38	151	206	192	<25
Zn	74	65	83	53	26	<25

**Figure A 6:** Traditional local kiln.  
Left: side view. Right: front view



**Figure A 7:** Covering of the kiln.  
Left: front is sealed with bricks and plaster. Middle: covering of the kiln before firing. Right: after firing.



**Figure A 8:** Advanced kiln in Kasongan which could be restored and used for filter production



**Figure A 9:** Pots in the traditional kiln before firing. Kiln is still open. Greenish process temperature control rings are distributed on each side on top of the pots and at the bottom of the kiln.





**Figure A 10:** Ceramic filter pots made of 40 % sawdust and 40 % rice husks.



**Figure A 11:** Surfaces of ceramic filter pots made of 0, 20 and 40 % saw dust.



**Figure A 12:** Experimental setup of filter experiments.



**Table A 4:** complete data for physico-chemical analysis of experiments with filters from local material Tap water (TW) and filtrates (F) of a filter containing 40 % sawdust (as in Table 12). (Mercury was <0.0001 mg/L, Antimony <0.001 mg/L, Iron <0.01 mg/L in all samples)

parameter	TW	F1	F2	F3+4	F5+6	F8	F10	F12	F14	F16	F18
pH [-]	7.39	7.75	8.24	8.23	8.27	8.32	8.32	8.33	8.17	8.56	8.35
EC [ $\mu$ S/cm]	716	873	662	629	638	666	688	691	705	729	701
Turbidity [NTU]	0.15	3.2	0.76	0.8	0.35	0.79	0.54	0.18	0.24	0.18	0.38
Boron [mg/L]	0.03	0.27	0.08	0.1	0.09	0.05	0.07	0.05	0.06	0.08	0.07
Cyanide [mg/L]	<0.01	<0.005	0.006	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Fluoride [mg/L]	0.08	0.91	0.75	0.5	0.42	0.37	0.47	0.59	0.69	0.53	0.49
Chloride [mg/L]	35.2	41.3	38	36.6	36.5	36.7	36.9	36.9	38.1	40.7	37.6
Sulfate [mg/L]	77.4	267	114	88.9	85.9	81.5	81.6	80.8	83.1	89	82.1
Ammonium [mg/L]	<0.01	0.36	0.06	0.05	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Nitrate NO <sub>3</sub> <sup>-</sup> [mg/L]	2.5	2.1	2.1	2.1	2	2	1.8	2.1	1.9	2.2	1.9
Nitrite NO <sub>2</sub> <sup>-</sup> [mg/L]	<0.01	0.14	0.04	0.01	0.04	0.01	0.01	<0.01	0.02	0.07	0.03
o-Phosphate [mg/L]	-	0.75	0.73	0.53	0.43	0.56	0.7	0.82	1.35	1.45	1.35
Total hardness [°dH]	19.0	17.9	8.8	15.9	15.8	17.5	18.6	18.9	18.8	20.8	18.9
Carbonate hardness [°dH]	-	5.7	9	10.1	10.3	12	12.8	13	13	14.1	13.4
Hardness [mg/L CaCO <sub>3</sub> ]	3.4	3.19	2.64	2.83	2.82	3.12	3.31	3.37	3.34	3.71	3.37

Acid capacity at pH 4.3 [mmol/L]	4.89	2.02	3.22	3.62	3.69	4.27	4.57	4.65	4.64	5.03	4.77
Oxidizability [mg/L]	-	2.1	1.54	0.57	0.44	0.44	0.53	0.53	0.4	0.36	0.61
Sodium [mg/L]	16.6	30.7	19.3	15.8	15.9	16.6	16.3	16.7	16.3	16.7	16.4
Potassium [mg/L]	3.5	21.8	10.2	8.55	4.69	3.8	3.32	3.33	3.19	3.28	3.21
Magnesium [mg/L]	14.7	12.1	10.6	11.4	12.2	13.2	13.6	13.9	14	15.2	14.1
Calcium [mg/L]	112	107.7	96.2	93.1	93	103.1	110.5	111.9	112.4	123.4	111.1
Calcite solubility [mg/L]	-17	-4.73	-24.86	-29.63	-31.38	-42.91	-49.24	-51.16	-47.14	-67.54	-53.22
Aluminum [mg/L]	<0.02	<0.005	0.014	0.005	<0.005	0.006	<0.005	<0.005	0.007	0.007	0.008
Arsenic [mg/L]	-	0.0006	0.036	0.024	0.022	0.021	0.02	0.019	0.021	0.021	0.015
Lead [mg/L]	-	0.003	0.003	0.002	0.004	0.002	0.001	0.002	0.002	0.002	0.002
Cadmium [mg/L]	-	<0.0001	0.0003	0.0002	0.0002	0.0002	0.0001	0.0001	0.0005	<0.0001	0.0004
Chromium [mg/L]	<0.001	<0.001	0.006	0.002	0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Copper [mg/L]	-	<0.001	<0.001	<0.001	0.002	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Manganese [mg/L]	<0.005	< 0.01	0.18	0.15	0.14	0.091	0.031	0.016	< 0.01	< 0.01	< 0.01
Nickel [mg/L]	-	<0.001	<0.001	<0.001	0.001	0.001	<0.001	0.002	0.002	0.001	0.001
Selenium [mg/L]	<0.001	<0.001	0.006	0.004	0.003	0.002	0.002	0.001	<0.001	0.002	<0.001
TOC [mg/L]	0.56	5.3	0.76	0.78	1.1	0.68	0.77	0.47	0.71	0.69	0.69
absorption at 436 nm [1/m]	<0.1	0.33	0.15	0.08	0.07	0.06	0.06	0.06	0.06	0.08	0.06
absorption at 254 nm [1/m]	0.9	62.5	23.5	14.4	13.9	13.1	11.6	9.6	8.8	7.2	5.4

**Table A 5:** Tap water (TW) and second filtrates F2 (F) of filter pots containing rice husks (R) and sawdust (S) in different concentrations (as in Table 13).

(F20 Ag = filtrate of filter made of 20 % sawdust coated with silver nitrate.

Mercury was &lt;0.0001 mg/L, Antimony &lt;0.001 mg/L, Iron &lt;0.01 mg/L in all samples)

parameter	TW	F40 R	F20 R	F0 R	F20 S	F20 Ag
pH [-]	7.39	8.68	-	7.98	8.38	8.52
EC [ $\mu$ S/cm]	716	658	-	445	587	566
Turbidity [NTU]	0.15	0.17	0.44	0.6	0.07	0.39
Boron [mg/L]	0.03	0.07	0.04	0.07	0.07	0.05
Cyanide [mg/L]	<0.01	<0.005	<0.005	<0.005	<0.005	<0.005
Fluoride [mg/L]	0.08	0.62	1.0	0.11	0.58	0.26
Chloride [mg/L]	35.2	36.7	36.6	37.4	42.0	37.0
Sulfate [mg/L]	77.4	86.0	81.0	84.7	98.3	93.5
Ammonium [mg/L]	<0.01	0.11	0.07	0.01	0.03	<0.01
Nitrate NO <sub>3</sub> <sup>-</sup> [mg/L]	2.5	1.9	2.0	1.9	1.7	2.7
Nitrite NO <sub>2</sub> <sup>-</sup> [mg/L]	<0.01	0.04	0.04	0.06	0.27	0.03
o-Phosphate [mg/L]	-	2.08	0.96	<0.03	0.24	0.11
Total hardness [°dH]	19.0	17.7	16.8	9.9	13.9	14.0
Carbonate hardness [°dH]	-	11.5	9.4	3.8	7.5	7.9
Hardness [mg/L CaCO <sub>3</sub> ]	3.4	3.15	2.99	1.77	2.47	2.49
Acid capacity at pH 4.3 [mmol/L]	4.89	4.09	3.37	1.37	2.68	2.82
Oxidizability [mg/L]	-	0.73	0.69	0.89	0.33	0.37
Sodium [mg/L]	16.6	18.3	17.1	15.7	16.4	16.1
Potassium [mg/L]	3.5	3.7	3.7	4.2	4.72	5.99
Magnesium [mg/L]	14.7	15.8	13.0	12.4	12.2	12.0
Calcium [mg/L]	112	99.9	87.1	50.3	81.9	79.9
Calcite solubility [mg/L]	-17	-43.3	-	0.444	-19.3	-23.4
Aluminum [mg/L]	<0.02	0.015	0.02	0.015	0.01	0.01
Arsenic [mg/L]	-	0.012	0.013	0.001	0.011	0.005
Lead [mg/L]	-	0.001	<0.001	0.001	0.001	0.002
Cadmium [mg/L]	-	0.0003	0.0002	0.0002	0.0005	<0,0001

Chromium [mg/L]	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Copper [mg/L]	-	0.008	0.007	0.003	<0.001	<0.001
Manganese [mg/L]	<0.005	0.47	0.44	0.72	<0.01	<0.01
Nickel [mg/L]	-	0.002	0.001	0.004	0.004	0.004
Selenium [mg/L]	<0.001	0.003	0.003	0.004	0.005	0.004
Silver [mg/L]	<0.003	-	-	-	<0.003	0.014
TOC [mg/L]	0.56	0.85	0.89	1.2	0.82	1.6
absorption at 436 nm [1/m]	<0.1	0.02	0.04	0.07	0.05	0.08
absorption at 254 nm [1/m]	0.9	6.74	10.86	9.43	13.9	12.3

**Table A 6:** Micro biological analysis of experiments with natural brook (BW) and river (RW) water and respective filtrates.

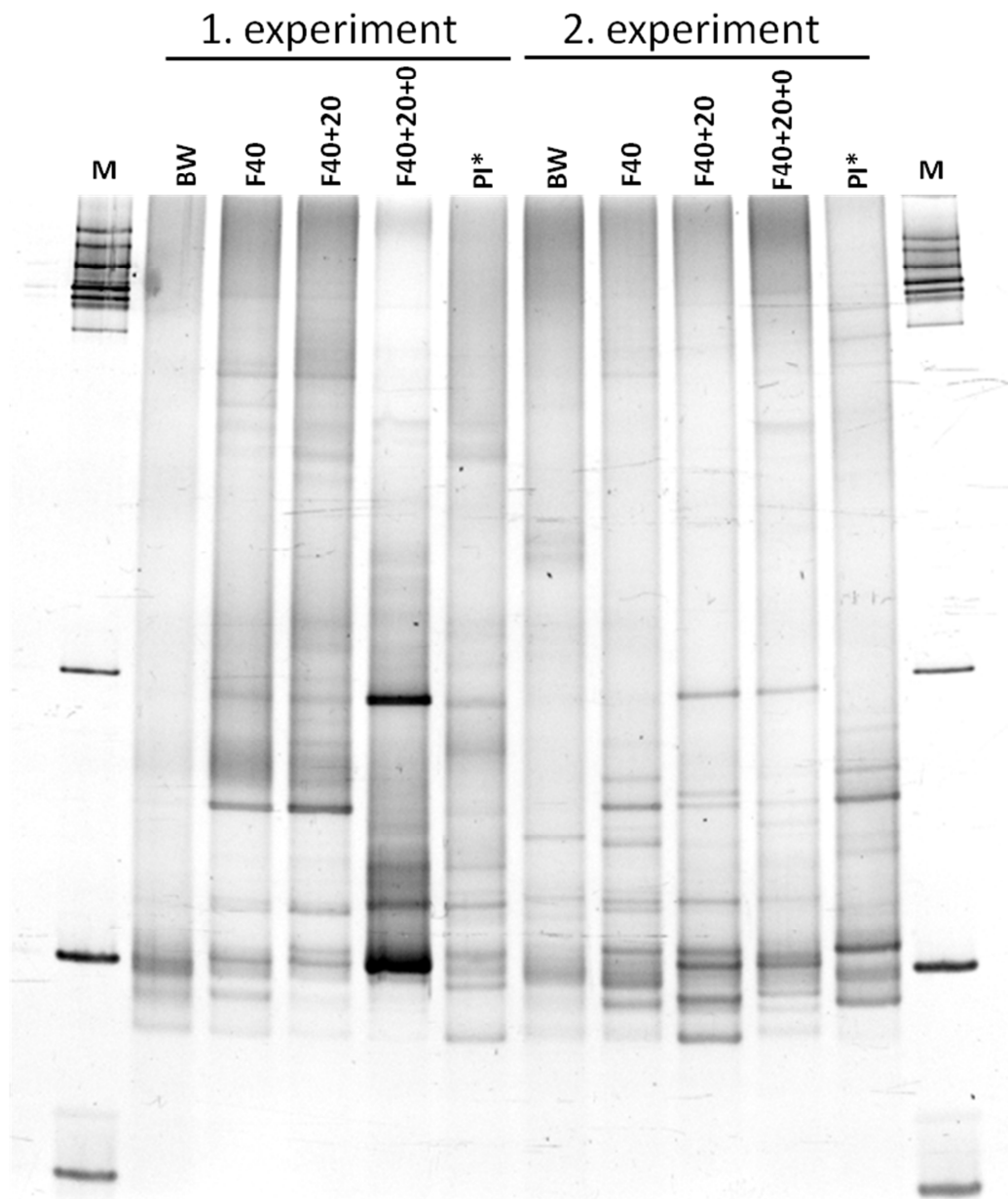
(Filtrates (F) of filter pots made of rice husks (R) and sawdust (S) in different concentrations, F40+20 = F40 filtered through filter with 20 % combustibles, F40+20+0 = F20 filtered through filter without combustibles, F20 Ag = F of filter made of 20 % sawdust coated with silver nitrate.

+++ many colonies ++ few colonies + very few colonies (+) 1-2 colonies – no colonies)

	HPC at 22 °C [CFU/mL]	HPC at 36 °C [CFU/mL]	Coliform bacteria [CFU/100 mL]	Enterococci [CFU/100 mL]
<b>Rice husk filters</b>				
Brook 1	BW	6.0*10 <sup>3</sup>	1.2*10 <sup>3</sup>	+++
	F40 R	3.6*10 <sup>4</sup>	1.3*10 <sup>4</sup>	++
	F40+20 R	2.8*10 <sup>4</sup>	1.8*10 <sup>4</sup>	++
	F40+20+0 R	2.1*10 <sup>5</sup>	4.4*10 <sup>4</sup>	+
Repetition	BW	5.5*10 <sup>3</sup>	1.2*10 <sup>3</sup>	++
	F40 R	2.7*10 <sup>4</sup>	3.9*10 <sup>3</sup>	++
	F40+20 R	8.9*10 <sup>4</sup>	1.4*10 <sup>4</sup>	++
	F40+20+0 R	5.1*10 <sup>4</sup>	1.0*10 <sup>4</sup>	+
River 1	RW	7.2*10 <sup>3</sup>	8.5*10 <sup>3</sup>	+++
	F40 R	1.1*10 <sup>4</sup>	2.5*10 <sup>3</sup>	++
	F40+20 R	2.6*10 <sup>4</sup>	3.4*10 <sup>3</sup>	++
	F40+20+0 R	2.4*10 <sup>5</sup>	1.5*10 <sup>4</sup>	+

Repetition	RW	$1.4 \cdot 10^4$	$3.9 \cdot 10^3$	+++	++	
	F40 R	$2.1 \cdot 10^4$	$3.9 \cdot 10^3$	++	(+)	
	F40+20 R	$2.1 \cdot 10^4$	$3.0 \cdot 10^4$	++	+	
	F40+20+0 R	$9.1 \cdot 10^4$	$2.7 \cdot 10^4$	+	-	
<b>Sawdust filters</b>						
Brook 1	BW	$7.6 \cdot 10^3$	$6.0 \cdot 10^2$	+++	++	
	F40 S	$4.5 \cdot 10^5$	$2.0 \cdot 10^5$	++	+	
	F20 S	$1.9 \cdot 10^3$	10	++	-	
	F0	$2.7 \cdot 10^5$	$7.5 \cdot 10^4$	+	(+)	
	F40+20 S	$2.7 \cdot 10^5$	$1.5 \cdot 10^5$	+	-	
	F40+20+0 S	$4.8 \cdot 10^5$	$3.1 \cdot 10^5$	+	-	
	F20 Ag	2	1	-	-	
	F40+20 Ag	$3.7 \cdot 10^3$	1	-	-	
	Repetition	BW	$2.4 \cdot 10^3$	$<1 \cdot 10^2$	+++	+
		F40 S	$1.0 \cdot 10^5$	$1.2 \cdot 10^5$		-
F20 S		$2.0 \cdot 10^5$	$1.4 \cdot 10^5$		-	
F0		$7.4 \cdot 10^4$	$4.5 \cdot 10^3$		-	
F40+20 S		$1.2 \cdot 10^4$	$<1 \cdot 10^2$	-	-	
F40+20+0 S		$1.4 \cdot 10^5$	$8.5 \cdot 10^3$	(+)	-	
F20 Ag		$2.1 \cdot 10^3$	14		-	
F40+20 Ag	$4.6 \cdot 10^3$	59	-	-		

**Figure A 13:** DGGE pattern of brook water samples filtered through rice husk filters. (BW = brook water, F40 = BW filtered through filter with 40 %rice husks, F40+20 = F40 filtered through filter with 20 % rice husks, F40+20+0 = F20 filtered through filter containing no rice husks.\*R PI represents the filtrate of the ceramic filter produced by Pelita.)





**Aspects that should be considered for local production of ceramic filters using locally available material**

- If possible, use of appropriate size of clay source to enable reproducible quality
- Screening of combustible material using two sieves to attain a more homogenous grain size distribution and thus a reproducible quality with finer pore sizes
- Mix components of traditional clay mixture (clay sources and lava sand)
- Addition of combustible material after mixture with water
- Mix clay with combustible material in ratio of about 80 % clay mixture : 20 % combustible material
- Addition of water and further adjustment of composition while mixing
- Sufficient drying depending on the dry / rainy season
- If possible, use of optimized kiln with good insulation
- Heating and cooling rate should be about 5 to max. 10 K/min in order to prevent cracks
- Firing temperature should be about 750 to 800 K; if higher temperature is used, concentration of pore-forming agents should be adjusted
- Coating with silver solution (silver nitrate or colloidal silver, depending on availability) by soaking in silver solution bath
- Extensive training of potters (including safety measures when handling silver solutions)

**Aspects that should be considered for the distribution of locally produced ceramic filters**

- Raising awareness for need of water treatment
- Extensive training of filter users in daily handling and maintenance of the filters (especially raising awareness for hygiene and possible recontamination of filter or filtered water)
- Providing long-term assistance
- Providing possibilities for water quality analysis

**Beberapa aspek yang harus diperhatikan dalam memproduksi filter keramik menggunakan material lokal**

- Jika memungkinkan, gunakan sumber lempung yang cukup agar memungkinkan untuk reproduksi dengan kualitas yang seragam
- Menyaring campuran bahan tambahan yang terbakar/pembentuk pori-pori (sekam padi) dengan dua lapis saringan agar ukuran butirnya lebih seragam dan menghasilkan kualitas yang memiliki ukuran pori-pori yang seragam dan halus.
- Campuran bahan tradisional sesuai yang sudah ada/biasa dibuat (lempung dan pasir halus/pasir lava)
- Penambahan bahan yang terbakar dengan perbandingan campuran lempung 80% dan bahan terbakar 20%
- Penambahan air sesuai keperluan saat pencampuran/pengadukan
- Pengeringan yang cukup sesuai dengan kondisi musim penghujan/kemarau
- Gunakan tungku pembakar keramik yang dengan teknologi yang memadai dan memiliki insulasi panas yang bagus
- Pemanasan dan pendinginan harus dalam kisaran 5 hingga maksimal 10 K/menit untuk menghindari retakan/pecah
- Suhu pembakaran harus dalam kisaran 750K hingga 800 K; jika suhu lebih tinggi dari itu, maka konsentrasi bahan pembentuk pori harus disesuaikan
- Pengecatan dengan cairan perak (silver nitrat atau colloidal silver tergantung yang ada) dengan merendam pada wadah larutan perak
- Pelatihan harus sering dilakukan untuk para pengrajin keramik (termasuk upaya keselamatan dalam menggunakan cairan perak)

**Beberapa aspek yang harus diperhatikan untuk distribusi filter keramik yang diproduksi lokal**

- Peningkatan kesadaran tentang pengolahan air
- Pelatihan penggunaan filter untuk keperluan harian dan perawatannya (khususnya meningkatkan kesadaran dan pengetahuan tentang kebersihan, higienitas dan kemungkinan kontaminasi filter dan air yang sudah difilter)
- Menyediakan pendampingan dalam jangka panjang
- Menyediakan akses untuk melakukan analisis kualitas air