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Multiple treatment objectives of solar driven electrolytic oxidant production for decentralized water treatment in developing regions and its economic feasibility

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Abstract

In 2017 nearly 2 billion people consumed water that was contaminated with feces, causing almost 500.000 diarrheal deaths. At the same time freshwater resources are depleted and water scarcity is already affecting 4 billion people worldwide. From a technical perspective the continuous supply of chemicals needed to ensure sufficient disinfection remains a major challenge in rural water treatment, and existing technical solutions to adequately disinfect water have failed in the past.

This dissertation work evaluates the technical and economic feasibility of solar-driven inline electrolytic production of chlorine (ECl_2), as an alternative to external chlorine supply. During ECl_2 disinfection the water passes through the cell and chlorine is produced “inline” from the natural chloride content of the water. Under optimal conditions, no chemicals are required to safely disinfect drinking water and treated wastewater. Furthermore, the ability of ECl_2 to enhance the removal of iron and arsenic from contaminated groundwater and to degrade Trace Organic Compounds (TOrC) when combined with UV were analyzed. All relevant tests have been conducted in long-term field studies in future deployment areas. This enabled the evaluation of potential operational challenges of such systems under real-world conditions. The experiences gathered from these field trials represent the major benefit of this dissertation work.

The trials have shown that with ECl_2 water can be safely disinfected and supplied with an adequate amount of residual disinfectant. Here, the combination with natural pretreatment systems has proven to be beneficial. For the drinking water trial conducted in Uttarakhand, India, the ECl_2 system received bank filtrate and achieved overall log removal rates of >5.0 for total coliforms and >3.5 for *E. coli*.

For the disinfection of treated wastewater, the combination with a vertical flow constructed wetland (VFCW) has largely simplified the disinfection with chlorine by equalizing wastewater (WW) inlet quality fluctuations, removing ammonium, COD, and turbidity. This has also substantially reduced the chlorine demand of the water, and pathogen indicator-free conditions were achieved with log unit removals of 5.1 and ≥ 4.0 for total coliforms and *E. coli*, respectively. Wastewater reuse applications that go beyond irrigation become permissible through this approach and the use of limited freshwater resources can be substituted.

Removal rates for arsenic and iron of 94 % and >99 % respectively were able to be achieved by treating contaminated groundwater with the combination of ECl_2 and subsequent co-precipitation and filtration proposed here. Despite effluent concentrations up to 10 ± 4 µg/L for arsenic, strict WHO guideline values could not be met. Here further optimization requirements were identified.

The removal rate for benzotriazole of 5% through ECl_2 alone could be increased to 89 % when combined with UV lamps. Similar results were achieved for other selected TOrCs. Still, more advanced studies are required to understand the degradation process in detail, and to evaluate a potential increase in toxicity through the formation of transformation products.

The field trials have shown that ECl_2 as an innovative treatment technology is capable of safely disinfecting drinking water and treated wastewater. Its application also enhances the treatment of other contaminants evaluated. However, cathode scaling has been identified as the most critical technical issue - despite the use of polarity inversion. ECl_2 systems could only operate reliably in waters with total hardness value < 200 mg/L $CaCO_3$. As such concentrations are rare, the fields of application of ECl_2 are limited. This required other chlorination technologies as alternatives to the originally planned inline electrolysis.

An initial derivative of an ECl_2 system was also applied during a wastewater disinfection trial in Spain. In this setting the portion of water passing by the electrodes and therefore the quantities of scaling agents were reduced to between 4 and 23 % of the total water volume treated. With this approach, deposit formation was completely prevented and reliable, nearly maintenance-free operation was ensured. However, such onsite chlorine generation (OCG) units commonly require the addition of chloride. From the author's perspective and the experience collected during the field trials, the addition of NaCl is justifiable considering the increased reliability of system operation. OCG offers further advantages regarding process stability and energy demand. Lab studies have also shown that the formation of inorganic disinfection byproducts has not been an issue with OCG systems.

To evaluate the economic feasibility of the drinking water treatment systems tested in real-world scenarios, an ECl_2 treatment system operating in Egypt and two OCG units operating in Tanzania and Nepal were analyzed. The study shows that long-term operation and maintenance costs of such units can be covered. However, seed investment is required for the construction of the initial infrastructure. Once those costs are covered, the treatment approaches presented here can sustainably play an important role in reducing the number of people consuming contaminated water, especially in rural developing regions.

Kurzfassung

Im Jahr 2017 konsumierten knapp 2 Milliarden Menschen fäkalkontaminiertes Wasser. Das führte zu fast 500.000 Todesfällen. Gleichzeitig werden Trinkwasserressourcen ausgebeutet und schon heute sind 4 Milliarden Menschen von Wasserknappheit betroffen. In ländlichen Entwicklungsregionen stellt, aus technischer Sicht, die Desinfektion des Wassers eine der größten Herausforderungen für die Sicherstellung einer angemessenen Trinkwasserversorgung dar.

In dieser Dissertation wird die technische sowie wirtschaftliche Machbarkeit einer solar betriebenen Anlage zur Chlorproduktion mittels Inline-Elektrolyse (ECl_2) als Alternative zur Lieferung und Dosierung von Chlorreagenzien analysiert und bewertet. Während der ECl_2 wird das gesamte aufzubereitende Wasser durch eine Elektrolysezelle geleitet und das Chlor aus dem natürlichen Chloridgehalt des Wassers „inline“ gebildet. Unter optimalen Betriebsbedingungen kann Trinkwasser aber auch aufbereitetes Abwasser ohne Zugabe durch Chemikalien desinfiziert werden. Damit würde die Lieferung von Chlorlösung dauerhaft entfallen. Zusätzlich wurde bewertet, inwiefern die Entfernung von Eisen und Arsen durch die ECl_2 sowie der Abbau von organischen Spurenstoffen durch zusätzliche Kombination mit UV-Bestrahlung zur Radikalbildung verbessert werden kann. Alle vorgestellten Feldtests wurden in Langzeitstudien unter Realbedingungen in zukünftigen Anwendungsgebieten durchgeführt. Dadurch konnten mögliche Probleme im Betrieb der Anlagen frühzeitig erkannt und Lösungsvorschläge erarbeitet werden. Die Erfahrungen aus dem Betrieb der ECl_2-Systeme stellen dabei den größten Nutzen dieser Arbeit dar.

Mit den Feldversuchen konnte gezeigt werden, dass Wasser durch die Anwendung von ECl_2 sicher desinfiziert und gleichzeitig ausreichend vor Wiederverkeimung geschützt werden kann. Insbesondere die Kombination mit naturnahen Verfahren zur Vorbehandlung des zu desinfizierenden Wassers hat sich als sehr vorteilhaft für die langfristig sichere Anwendung des Verfahrens herausgestellt. Für den in Uttarakhand, Indien, durchgeführten Feldtest konnte die ECl_2 zusammen mit der Uferfiltration eine Logstufenreduktion von > 5.0 für Gesamtcoliforme und 3.5 für *E. coli* erreichen.

Durch die Kombination mit einer vertikal durchströmten Pflanzenkläranlage (VFCW) konnte die Desinfektion von behandeltem Abwasser mittels Chlor erheblich vereinfacht werden. Die Pflanzenkläranlage reduzierte die Ammoniumkonzentration, den CSB sowie die Trübung und damit auch die Chlorzehrung des Wassers. Darüber hinaus wurden durch die VFCW Konzentrationsschwankungen im Zulauf erheblich vergleichmäßigt. Es wurden Logstufenreduktionen für Gesamtcoliforme von 5.1 und >4.0 für *E. coli* erzielt. Durch diese Verfahrenskombination werden auch Anwendungen zur Wiederverwendung von Abwasser möglich, die über die Bewässerung hinausgehen und damit wertvolle Frischwasserressourcen geschont.

Durch die Behandlung von arsenkontaminiertem Grundwasser konnten mit der hier vorgeschlagenen Kombination von ECl_2 und anschließender gemeinsamer Ausfällung und Filtration Entfernungsraten für Arsen von 94 % und Eisen von > 99 % erreicht werden. Da der WHO-Grenzwert für Arsen (10 µg/L) im hier durchgeführten Feldversuch mit 10 ± 4 µg/L dauerhaft nicht sicher eingehalten werden konnte, wurden weitere Optimierungsschritte identifiziert.

Die Entfernungsrate für Benzotriazol von 5 % durch ECl_2 allein konnte in Kombination mit UV-Lampen auf 89 % erhöht werden. Ähnliche Ergebnisse wurden für andere ausgewählte organische Spurenstoffe erzielt. Es sind jedoch weitere Studien erforderlich, um den Abbauprozess im Detail zu verstehen und eine mögliche Zunahme der Toxizität durch die Bildung von Transformationsprodukten sowie Desinfektionsnebenprodukten zu bewerten.

Die Feldversuche haben gezeigt, dass ECl_2 als innovative Behandlungstechnologie in der Lage ist, Trinkwasser und behandeltes Abwasser sicher zu desinfizieren. Darüber kann mittels ECl_2 u.a. die Entfernung von Arsen aus verunreinigten Rohwässern als auch der Abbau von Spurenstoffen verbessert werden.

Bei härterem Rohwasser steht die rasche Verkalkung der Kathoden jedoch einem wartungsarmen Betrieb der Anlagen, trotz Polumkehr, entgegen. Die Versuche haben gezeigt, dass ECl_2-Systeme mit den hier verwendeten Elektrolysezellen, nur in Wässern mit einem Gesamthärtewert <200 mg/L $CaCO_3$ zuverlässig arbeiten. Da Rohwässer häufig Konzentrationen >200 mg/L aufweisen, ist der Anwendungsbereich der ECl_2 begrenzt und erfordert alternative Chlorierungstechnologien zur ursprünglich geplanten Inline-Elektrolyse.

Bereits während Versuchen zur Abwasserdesinfektion in Spanien wurde daher das Pilotsystem technisch dahingehend angepasst, dass nur noch ein Teilstrom von 4 bis 23 % des zu behandelten Wasservolumens durch die Elektrode floss. Dadurch konnte die Bildung von Ablagerungen vollständig verhindert und ein zuverlässiger, nahezu wartungsfreier Betrieb sichergestellt werden. Je nach dem Chlorbedarf und der natürlichen Chloridkonzentration des Wassers erfordert diese Betriebsweise in der Regel jedoch die Zugabe von Chlorid. In Anbetracht der hier ermittelten erhöhten Prozessstabilität und dem erheblich reduzierten Energieverbrauch erscheint diese Zugabe vertretbar. Laborstudien haben auch gezeigt, dass die Bildung anorganischer Desinfektionsnebenprodukte bei den „onsite chlorine generation“ (OCG) Systemen kein Problem darstellt.

Um die wirtschaftliche Machbarkeit der hier getesteten Trinkwasseraufbereitungssysteme unter Realbedingungen zu bewerten, wurden der Betrieb eines in Ägypten eingesetzten ECl_2-Aufbereitungssystems und zweier in Tansania und Nepal eingesetzter OCG-Einheiten analysiert. Die Studie zeigt, dass die Betriebs- und Wartungskosten solcher Einheiten dauerhaft gedeckt werden können. Für den Aufbau der Infrastruktur sind jedoch Investitionen durch entsprechende Förderprogramme erforderlich.

Die hier angewandten Verfahren zur Wasseraufbereitung können eine wichtige Rolle bei der Verbesserung der Trinkwasserversorgung insbesondere in ländlichen Entwicklungsregionen und der Wiederverwendung von aufbereiteten Abwässern spielen.

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Philipp Otter Kassel, April 2021

Table of content

Abbreviations

AFM – Activated Filtration Media

AOP – Advanced Oxidation Process

BEP – Break Even Point

BDD – Boron Doped Diamond electrodes

CAC – Combined Available Chlorine

COD – Chemical Oxygen Demand

CSB – Chemischer Sauerstoffbedarf

CW – Constructed Wetland

DBPs – Disinfection By-products

DC – Direct Current

DOC – Dissolved Organic Carbon

DSA – Dimension Stable Anodes

DW – Drinking Water

ECl_2 – Inline Electrolytic Production of Chlorine

ET – Evapotranspiration

FAC – Free Available Chlorine

GSP – Greensand Plus

GW – Groundwater

HAAs – Haloacetic Acids

HFCW – Horizontal Flow Constructed Wetland

HNM – Halonitromethanes

HRT – Hydraulic Retention Time

LoD – Limit of Detection

MBR – Membrane Bioreactor

MDG – Millennium Development Goals

MENA – Middle East and North Africa

MOX – Mixed Oxide Electrode

MP – Madhya Pradesh

NDMA - N-Nitrosodimethylamine

NOM – Natural Organic Matter

NFD – Niederfraundorf

NPV – Net Present Value

OCG – Onsite Chlorine Generation

ORP – Oxidation Reduction Potential

O&M – Operation and Maintenance

PE – Person Equivalent

PV – Photovoltaic

PrV – Present Value

RBF – Riverbank Filtration

SDG – Sustainable Development Goals

THMs – Trihalomethanes

TOC – Total Organic Carbon

TOrC – Trace Organic Components

TSS – Total Suspended Solids

TZW – Technologiezentrum Wasser (German Water Centre)

UK – Uttarakhand

UP – Uttar Pradesh

WB – West Bengal

WW – Wastewater

WWTP – Wastewater Treatment Plant

VFCW – Vertical Flow Constructed Wetland

VFW – Vertical Filter Wells

1 Introduction

1.1 Background

The Sustainable Development Goal (SDG) No. 6 claims to “Ensure access to water and sanitation for all” by 2030. However, still today worldwide nearly 800 million people have no access to an improved drinking water (DW) source and between 1.8 and 2 billion people consume drinking water, that is classified as improved, but contains fecal coliforms (Onda *et al.* 2012; Bain *et al.* 2014b; Bain *et al.* 2014a). The consumption of such waters causes 500,000 deaths per year as a result of diarrheal diseases (WHO 2019). Hereby water in rural areas is more often contaminated than in urban areas, with Africa and South-East Asia suffering more than other continents (Bain *et al.* 2014a). In the past, community or centralized piped drinking water supply interventions focused rather on the availability of water in sufficient quantities than on its quality (LeChevallier and Au 2004). Emphasis on supplying water at the required quality over simply supplying access in rural communities has only started to gain importance in 2001 (Clasen and Cairncross 2004; Kremer 2007). Only with the SDGs the global efforts to achieve defined levels of water quality have become a matter of concern in 2015. Water treatment should, prior to all potential other contaminants, maintain microbial quality requirements (WHO 2017). In the existing legal drinking water frameworks, the absence of indicator pathogens is urgently required after treatment, during distribution and consumption of the water. To achieve this, the presence of residual disinfectant in the water is usually mandatory to assure protection against recontamination. Depending on the source water qualities additional treatment steps are necessary to meet existing guideline values. Those range from the simple removal of particulate matter allowing safe disinfection and long-term operation of disinfection systems to complex treatment requirements for the removal of e.g., arsenic and - with growing importance - trace organic components (TOrCs) from water sources.

Next to the drinking water quality challenges, increasing population growth, with an expected population of up to 9.7 billion inhabitants by 2050 (UN 2019a), uneven distribution of water resources, and climate change limit water availability and increases water stress in many regions worldwide. Already today about 4 billion people suffer from severe water scarcity during at least one month of the year (Mekonnen and Hoekstra 2016). One third of the world’s biggest groundwater systems are already in distress (Richey *et al.* 2015), being a potential driver of regional insecurity and water scarcity is a major challenge for socio-economic development (World Bank 2017). 17 countries, comprising 25 % of the world population, face “extremely high” levels of water stress. Here, agriculture, industries and municipalities consume more than 80 % of the average annual water supply, leaving these areas very vulnerable towards fluctuations caused e.g. by droughts and/or increased water withdrawals (Hofste *et al.* 2019). With 12 out of these 17 countries the Middle East and Northern Africa region (MENA) is mostly affected by water scarcity (World Bank 2017). Although most population growth will occur in small towns and cities the population in rural areas will also increase - in rural Africa by 43 % until 2050, which equals about 900 million additional consumers (UN 2019b). Increasing water stress, source water quality deterioration as well as population growth in developing areas require continuous efforts and new approaches for meeting the water quantity and quality requirements in rural regions. Here the reuse of treated wastewater (WW) for e.g. irrigation and process water applications has become an important part of an integrated water resource management maintaining or even increasing the availability of drinking water sources.

The application of chlorine as disinfectant is usually required to meet the legislative requirements for drinking water supply as well as for the safe reuse of WW (EPA 2012; WHO 2017). Chlorine is a highly effective disinfecting agent against many enteric bacteria, virus, spores, and protozoa (Tyrell et al., 1995; Sobsey et al., 1998; Veschetti et al., 2003). The major advantage, however, against alternative disinfection technologies, such as membrane filtration or UV radiation is that chlorination has a long proven record of keeping water safe during storage and distribution - if handled correctly (Clasen *et al.* 2007; Hashmi *et al.* 2009).

In rural communities water treatment systems based on chlorination have failed due to the requirements of a constant availability of chemicals, skilled personnel capable in correct handling and dosing as well as strict compliance with existing guidelines (Jaskolski *et al.* 2019). Often the implementation of existing technologies, that work well in more centralized settings, has failed in rural areas due to many reasons. Among them, the most prevalent ones are the lack of technical skills, inadequate system design, as well as the requirements for a constant supply, storage and handling of chemicals, have been identified (Jaskolski and Pellegrino 2019). For the removal of e.g., heavy metals, such as arsenic, a variety of adsorption media are available. Those work well in principle, but the success of their application depends on a community’s long-term ability to exchange such media. Lacking financial resources and the unavailability of high quality media have prevented communities to comply with such requirements and are the main reason for system failures as e.g. shown by Hossain *et al.* (2005), who examined the efficiency of arsenic removal plants in West Bengal, India.

Therefore, site-specific water treatment approaches tailored to the water and consumer requirements of rural developing areas need to be developed.

The inline electrolytic production of chlorine (ECl_2) has often been considered as potential disinfection technology to facilitate decentralized water treatment. Lab studies can be traced back to the 1950ies, 70ies, 80ies of the last and the 10er years of the recent century (Reis 1951, 1976; Kirmaier *et al.* 1980; Bergmann 2010; Schmidt 2012). The major advantage is seen in the production of chlorine from the natural chloride content of the water that is to be treated. ECl_2 hereby offers the appealing possibility to chemically disinfect water without the need for any external supply of chemicals. In former publications the effects of cell design, plate coatings, water temperature, chloride content on the chlorine production rate, disinfection-by-product (DBP) formation, salt yield, and the current efficiency have been examined (Kraft *et al.* 1999b; Kraft *et al.* 1999a; Bergmann and Koparal 2005; Schmidt 2012). However, such studies were conducted under lab conditions assuming that the water to be treated is clear and only requires disinfection. Data on long-term performance and operational challenges under realistic field operations of ECl_2-based treatment systems are not available or have not been published.

In real case treatment settings the disinfection is always the last step in water treatment and preceding steps are required to assure a successful application of ECl_2 systems. This increases complexity and costs and therefore challenges the application in rural settings.

Here the combination with natural (pre)-treatment systems, such as riverbank filtration (RBF) or constructed wetlands (CWs), offers the possibility to sustainably deal with a wider range of water quality issues. Pathogen loads, suspended solids, biodegradable organic matter and absorbable compounds of contaminated surface waters are substantially reduced during RBF at very marginal costs (Maeng 2010; Sandhu and Grischek 2012). CWs have proven to be a reliable alternative to more technical systems for the treatment of WW in rural settings - if the required space is available. In combination with such natural treatment approaches ECl_2 may pose a robust solution that can be operated on the long-term to supply safe water.

Additional to that, the production of chlorine as strong oxidizing agent may allow the development of more innovative treatment steps, that enhance the removal of other contaminants. Elevated concentrations of iron, arsenic as well as TOrCs have been identified as potential candidates, which's removal can be enhanced by the application of ECl_2. Especially if the required electricity can be supplied by solar systems, ECl_2 may pose a sustainable treatment solution to meet the drinking and WW treatment requirements in remote rural developing areas.

To assure long-term operation and maintenance of such treatment systems adequate business models are required and the generation of income through the supply of water has proven to be of importance. In rural drinking water supply the implementation of water points and water kiosks play a key role in developing such business models (Foster 2017; Hope *et al.* 2020). Water points have, in combination with water volume-based tariffs and innovative mobile payment schemes, helped to reduce non-revenue water and increase the income generated (Foster 2017). However, data on the financial feasibility of ECl_2 based treatment systems in combination with water point-based drinking water supply in low-income settings are not available. Therefore, site specific long-term evaluations are required considering investment, operation, and maintenance (O&M) costs as well as revenue streams to evaluate the economic feasibility of such settings.

1.2 Motivation

As data on the applicability of ECl_2 and its economic feasibility under real field conditions are lacking scientifically accompanied pilot trials are required. Those pilot trials will allow to identify operational limitations especially when operated in rural and remote developing regions. The conduction and evaluation of such pilot trials form the basis of this dissertation work, and will focus on the generation of data and the evaluation of:

1. the disinfection performance of ECl_2 based drinking water treatment systems combined with a pilot RBF setting tested in Uttarakhand, India,
2. ECl_2 and its ability to enhance the removal of elevated iron and arsenic concentrations by conducting field tests in Germany, Costa Rica and West Bengal, India,
3. the combination of a constructed wetland with an Onsite Chlorine Generation (OCG) system and its ability to allow safe reuse of treated WW in remote settings, tested on a WW treatment plant in Almeria, Spain,
4. the potential of ECl_2 as part of an Advanced Oxidation Process (AOP) when combined with UV radiation to produce radicals and by that oxidize TOrCs tested in Dresden, Germany.

Whereas the drinking water treatment and supply systems are intended for safe community water supply, the target of the trial conducted in Spain was to evaluate the application of electro-chlorination for WW disinfection allowing the reuse of WW in rural regions. Due to hardness related operational challenges, a first derivation from the originally planned ECl_2 system towards an OCG system was required and the newly developed setting was tested in Spain.

Adequate financial management approaches are required to ensure long-term operation and maintenance of such systems. Therefore, this dissertation work will also evaluate the:

5. economic feasibility of three successfully installed treatment systems in Nepal, Egypt, and Tanzania.

The main objective of this dissertation work is to develop, test and prove technical settings that allow the operation of such systems under challenging environmental and socio-economic conditions found in low-income and rural developing regions. It is hypothesized that such systems:

a. are capable in sufficiently disinfecting the water and removing the above-mentioned contaminants to meet existing guideline values,
b. can operate with, at most, monthly personal maintenance checkups which can be conducted by local technicians,
c. do not require any external chemical supply (except NaCl) to operate,
d. do not require an external supply of electricity,

e. do not exceed existing guidelines with regards to disinfection by-products and
f. can cover long-term O&M costs when used in rural drinking water supply.

In case those hypotheses cannot be confirmed, limiting operating conditions challenging the application of ECl_2 will be defined and alternatives to the supply of chlorine through ECl_2 proposed. Besides the generation and scientific interpretation of data produced during lab and field work, the assessment of long-term field data and operational experiences gathered represent the major benefit of this dissertation work. Provided that water quality can be improved with ECl_2 based treatment, such systems could play an important role in reaching SDG No. 6.

1.3 Outline

This cumulative dissertation is a cumulative document including nine articles published.

In chapter 2 the applied analytical methodologies are presented. In chapter 3 the basic principles of chlorination in water disinfection and the production of chlorine by means of ECl_2 will be described. Key findings from literature and own lab studies show the dependencies of chlorine production rates and efficiencies on chloride concentration, current densities, pH, and hydraulic retention times of the here applied electrolytic cells. Further, the challenges in the operation of ECl_2 systems and source water characteristics limiting the application of ECl_2 as well as treatment alternatives will be pointed out.

In chapters 4-7 the above-described field trials are summarized, and the findings of each field test presented. In chapter 9 the economic feasibility of decentralized drinking water supply infrastructures, based on the here pilot tested technologies, is evaluated. The most important articles published in international scientific peer-reviewed journals are enclosed to this thesis. References to those publications are labelled by [Px] in the text, in Tab. 1, and are listed at the end of the chapters. In chapter 8 the potential advantages of OCG as an alternative approach for chlorine supply in decentralized settings will be described. In chapter 10 the major findings of the dissertation work will be summarized and future research aspects are formulated in chapter 11.

Tab. 1: Pilot sites and treatment targets of systems evaluated within this dissertation thesis

Pilot Sites	**Treatment perspective of pilot system**							**Publication***
	Reduction of					Disinfection of		
	Fe	As	Mn	Turbidity	TOrCs	DW	WW	
Evaluated for treatment performance								
NFD, Saxony, Germany	x	x	x	x		x		[P1.1]
San Jose, Costa Rica	x	x	x	x		x		[P1.2]
Kalyani, WB, India	x	x	x	x		x		[P1.3]
Haridwar, UK, India**				x		x		[P2.1]
Aligarh, UP, India				x			x	[P3.1]
Amarkantak, MP, India				x		x	x	[P3.2]
Almeria, Spain**				x		x	x	[P3.3]
Hosterwitz, Saxony, Germany**					x	x		[P5]
Evaluated for economic feasibility								
Rombo, Tanzania						x		[P4]
Devgaun, Nepal	x			x		x		
El Heiz, Egypt	x		x	x		x		

* numbering follows sequence of publication date ** also evaluated for org. and/or inorg. DBPs

2 Methods

As most pilot trials were conducted in real case test settings, sampling, preservation of samples and analysis was challenging. Samples were either analyzed onsite using handheld devices, or, wherever possible, in nearby labs using best available equipment. Due to the very different trial locations and settings of the case studies applied analytical methods are partly inhomogeneous, e.g. for the analysis of indicator pathogens. As laboratories with functioning and adequate analytical methods were generally not available the application of handheld photometers (e.g. AL410, Aqualytic Dortmund Germany) for the in-field analysis of physicochemical parameters has proven to be very efficient and was used throughout the different test sites. Only for the analysis of the samples collected in Niederfraundorf (NFD) [P1.1] inductively coupled plasma optical emission spectroscopy (ICP-OES) was available. The methods applied are described in the following.

As **pathogen indicators** total coliform and *E. coli* were analyzed by either using IDEXX© Colilert© Quanti-Tray© [P2.1], membrane filtration [P3.3 and P3.1], or MPN [P3.2] method. **Electric conductivity, temperature,** and **pH** were measured with WTW Multi 3430 [P1.1] and Hach HQ40d (all other sites). **Oxidation reduction potential** (ORP) was measured using WTW Multi 350i [P1.1] or HQ40d (all other sites). ORP was additionally measured through a pilot system integrated ORP setting using tecLine Rd electrode (Jumo, Fulda, Germany). **Turbidity** was determined using an Aqualytic turbidity meter AL250T-IR (Dortmund, Germany) [P3.3] and WTW Turb 430 IR/T from (Weilheim, Germany) [P2.1]. For parameters documented in Tab. 2 an AL410 handheld photometer (Aqualytic, Dortmund, Germany) was used.

Tab. 2: Analysis of physicochemical parameters

Parameter	Wavelength	Method	Range
Fe_{total} [mg/L]	530	1,10-Phenanthroline	0.02-3.00
Mn_{total} [mg/L]	560	PAN	0.01-0.70
PO_4-P [mg/L]	660	Phosphomolybdenum blue Ascorbic acid	0.06-2.50
NH_4-N [mg/L]	610	Indophenole	0.02-1.00
Cl^- [mg/L]	530	Silver nitrate / turbidity	0.5-25.0
Total Hardness in [mg/L] $CaCO_3$	560	Metalphthalein	2-50
Free Chlorine in [mg/L]	530	DPD1	0.01 – 6.00
Total Chlorine in [mg/L]	530	DPD3	0.01 – 6.00
Total Chlorine high range in [mg/L]	530	Potassium Iodine/acid	5-200

Total arsenic [P1.2 and P1.3] was measured with an Arsenator® digital arsenic test kit (Palintest, Gateshead, UK). During the pilot study in NFD [P1.1], the concentration of **cations** (As, Fe, Mn, Ca and Mg) was determined with inductively ICP-OES using an Optima 4300 DV by PerkinElmer (Waltham, USA). The concentration of **anions** ($H_2AsO_4^-$, Cl^-, SO_4^{2-}, NO_3^-, NO_2^-) was measured using an ion-exchange chromatograph ICS 900 by Dionex (Sunnyvale, USA).

Chemical oxygen demand (**COD)** was determined using DR3900 (Hach Lange, Düsseldorf, Germany) [P3.3] following DIN 38409-H41-H44. **TSS** was determined following APHA Method 2540 D.

Samples for DBP analyzes taken in India [P2.1] were transported to Germany and analyzed in Technologiezentrum Wasser (TZW) lab in Dresden. **Inorganic DBP** [P5 and P3.3] (chlorate, chlorite, perchlorate, bromate) were analyzed following DIN EN ISO 10304-4 and TZW lab method, using an ICS 3000 by Thermo Fischer Scientific (Waltham, MA, USA) having a detection limit of 1 µg/L. **Trihalomethanes** (THMs) [P2.1 and P5] were analyzed following DIN EN ISO 10301, using a 7890A GC/MS by Agilent Technologies (Santa Clara, CA, USA) with a detection limit of 0.1 µg/L. Samples containing chlorine were quenched using thiosulfate directly after sampling.

For the determination of specific **energy** requirements during electrolytic chlorine production and pumping, current, voltages and flow rates were measured by using the pilot system integrated SCADA systems during the pilot trials. During lab tests the values given by the power source (BaseTech BT-305, Hirschau, Germany) were recorded to determine the power consumption of the electrolytic cells. Total water volumes were recorded by calibrated water meters.

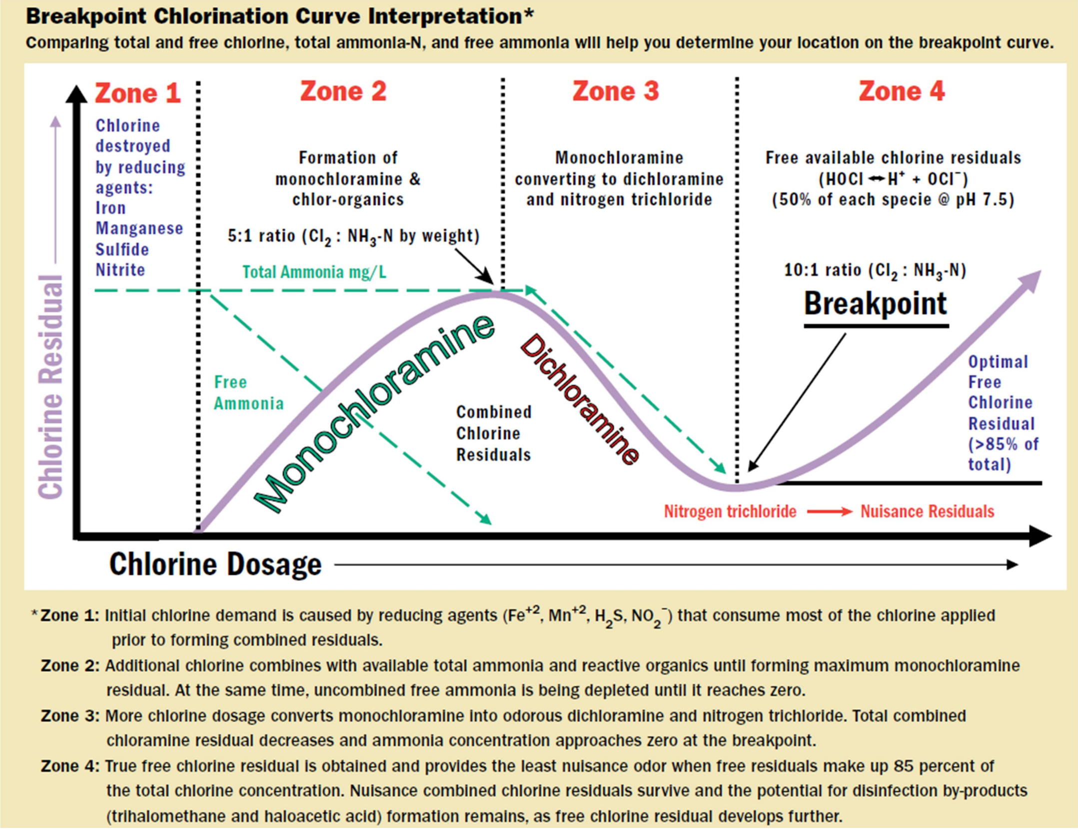
More detailed descriptions of applied analytical methods, along with derivations from the here described methodologies and additional site-specific parameters evaluated, are documented in the respective publications.

3 Chlorination in water disinfection

3.1 Chlorine as effective water disinfectant

Chlorination has a long record for safe drinking water disinfection. Next to its effectiveness against a wide range of potentially present pathogens its major advantage towards alternative disinfectants is the supply of residual protection from recontamination of the water during transport and storage.

Chlorine added into water will react with inorganic and organic materials creating a chlorine demand, which is specific for each water source and may change over time. Ammonium, often found in contaminated water sources, will react with chlorine via monochloramine, which is considered as combined available chlorine (CAC), to dichloramine and finally via nitrogen trichloride to nitrogen gas, marking the chlorination breakpoint (Fig. 1). Free Available Chlorine (FAC) will be available only after such reactions are completed.



Breakpoint Chlorination Curve Interpretation*

Comparing total and free chlorine, total ammonia-N, and free ammonia will help you determine your location on the breakpoint curve.

* **Zone 1:** Initial chlorine demand is caused by reducing agents (Fe^{+2}, Mn^{+2}, H_2S, NO_2^-) that consume most of the chlorine applied prior to forming combined residuals.

Zone 2: Additional chlorine combines with available total ammonia and reactive organics until forming maximum monochloramine residual. At the same time, uncombined free ammonia is being depleted until it reaches zero.

Zone 3: More chlorine dosage converts monochloramine into odorous dichloramine and nitrogen trichloride. Total combined chloramine residual decreases and ammonia concentration approaches zero at the breakpoint.

Zone 4: True free chlorine residual is obtained and provides the least nuisance odor when free residuals make up 85 percent of the total chlorine concentration. Nuisance combined chlorine residuals survive and the potential for disinfection by-products (trihalomethane and haloacetic acid) formation remains, as free chlorine residual develops further.

Fig. 1: Breakpoint chlorination and interpretation of the curve (Spon 2008)

Monochloramine itself is also considered as a residual disinfecting agent, with a largely reduced disinfection capacity and DBP-formation potential. Total chlorine (TC) is the sum of FAC and CAC (Eq. 1).

$$FAC + CAC = TC \qquad \text{(Eq. 1)}$$

Depending on the pH, FAC comprises chlorine gas (Cl_2) and hypochlorous acid (HOCl) as a strong oxidizing agent and its anion OCl^-. Due to the reduced oxidation capacity of OCl^- towards HOCl the application of chlorine is not suggested for pH values higher than 8.5 (Fig. 2).

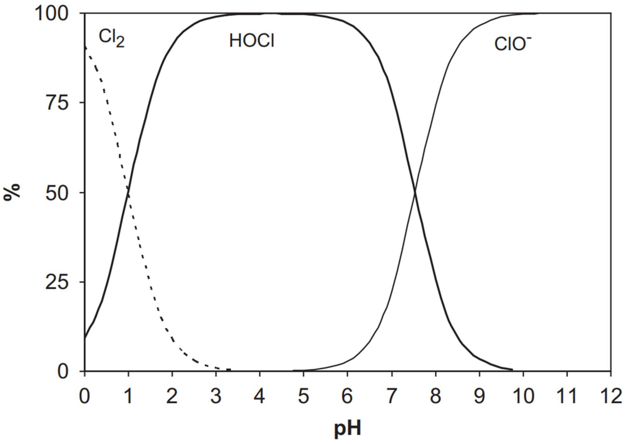


Fig. 2: Percentage distribution of Cl_2, HOCl and ClO^- as a function of pH at 25°C and chloride concentration of 177.5 mg/L (Deborde and Gunten 2008)

The application of chlorine for water disinfection is often questioned by the formation of unwanted disinfection DBPs. Among them the THMs and haloacetic acids (HAAs) are regulated due to their potential carcinogenicity and their limiting guideline values are given in Tab. 3. However, the risks associated with pathogenic contamination substantially exceed the adverse side effects of chlorination and “should not be compromised in attempting to control disinfection by-products” (WHO 2017) especially when considering the described drinking water supply situation in developing regions. The formation of DBPs should be monitored and reduced as much as possible. Hereby the focus needs to be set on using water with low total organic carbon (TOC) levels or on reducing such levels prior to disinfection.

In some countries the chloramination is considered as alternative to the application of FAC, as this is less prone to form THMs. This approach, however, has been questioned in the past due to the formation of N-Nitrosodimethylamine (NDMA) or halonitromethanes (HNMs) (Song *et al.* 2010; U.S. EPA 2014) and is banned in several European countries.

3.2 Inline electrolytic chlorine production

3.2.1 Basic reactions

The application of chlorine solution or chlorine gas are standard procedures in central water treatment systems. In remote rural developing regions, however, the supply, storage, handling, as well as the dosing is technically challenging and standard protocols are rarely followed. This leads to under- and overdosing of chlorine, resulting in a reduced disinfection rate, poor smell, and taste, as well as environmental issues and all such challenges have contributed to a negative image of chlorine in water treatment.

The electrolytic production of chlorine (electro-chlorination) (Fig. 3a and b) allows the production of chlorine directly at the site of treatment, through which transport and storage can be substituted. Here chloride ions are oxidized to chlorine gas at the anode as shown simplified in Eq. 2.

Anodic reaction chloride:

$$2Cl^- \leftrightarrow Cl_2 + 2e^- \qquad \text{(Eq. 2)}$$

Cathodic reaction:

$$2H_3O^+ + 2e^- \leftrightarrow H_2 + 2H_2O \qquad \text{(Eq. 3)}$$

Anodic side reaction oxygen:

$$2H_2O \leftrightarrow O_2 + 4H^+ + 4e^- \qquad \text{(Eq. 4)}$$

Hydrolysis of chlorine gas in water:

$$Cl_2 + 2H_2O \leftrightarrow HClO + Cl^- + H_3O^+ \qquad \text{(Eq. 5)}$$

The chlorine gas production is accompanied by a decrease of pH and the evolution of hydrogen gas at the cathode represents the required reduction reaction at the cathode (Eq. 3). The generation of oxygen at the anode is a side reaction (Eq. 4), reducing the efficiency of the process. Through the hydrolysis of chlorine gas in water hypochlorous acid is formed (Eq. 5), which is the actual disinfecting agent in water.

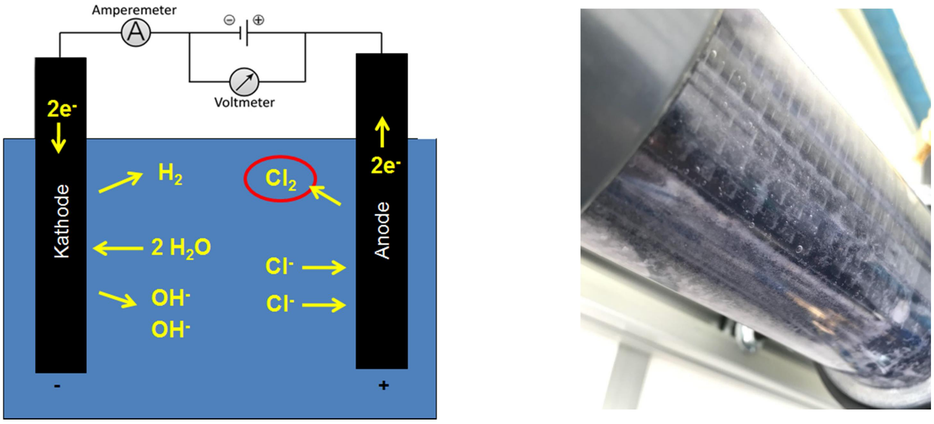


Fig. 3a and b: Scheme of chlorine electrolysis (a) and reactor used during the studies (b)

In this work the focus was originally set on the inline electrolytic production of chlorine (ECl_2) using electrodes coated with iridium and ruthenium oxides (MOX electrodes). Here the complete volume of the water to be treated passes the electrolytic chamber and the chlorine is produced directly from the natural chloride content of this water. Most source water to be disinfected already contains sufficient chloride to allow the production of chlorine in quantities required to safely disinfect the water. To power this process DC voltage is applied to the MOX electrodes. Studies have shown that MOX electrodes produce consistently higher chlorine output compared to platinum coatings (Kraft *et al.* 1999b). In comparison to boron doped diamond (BDD) electrodes described in literature, MOX electrodes are less prone to produce chlorate and perchlorate (Haaken *et al.* 2012; Schmidt 2012). At a later stage of this dissertation work the same electrodes were used to produce chlorine form a diluted NaCl brine solution through OCG, which follows the same chemical reactions given above.

The major influencing factors on the efficiency of the chlorine production through ECl_2 are described in detail in the following sections.

3.2.2 Chloride concentration

Kraft (2008) and Kraft *et al.* (1999b) show that the concentration of FAC increases with chloride concentration at a given current or current density. This was confirmed in own lab studies conducted with the here used MOX electrodes. At currents between 2 and 8 A (6.3-25.1 mA/cm²) the chloride concentration and produced FAC are linearly related as shown in Fig. 4a, considering chloride concentrations of 25, 50 and 100 mg/L.

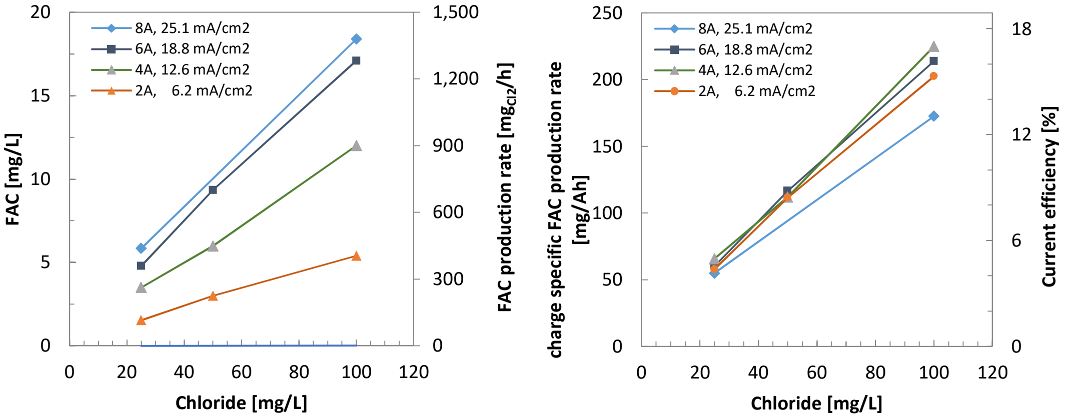


Fig. 4 a and b: Dependence of FAC concentration, FAC production rate (a), charge specific FAC production rate and current efficiency (b) on chloride concentration at the given currents (75 L/h, 400 µS/cm, pH=7, 18°C)

With the here applied MOX electrodes the achieved FAC production rates ranged from 0.12 g/h for 2 A and chloride concentration of 25 mg/L to 1.38 g/h for 8 A and chloride concentration of 100 mg/L. Fig. 4a and b also show that the effect of increasing the current density on the production rate is limited. Even though higher production rates can be reached, the charge specific FAC production rate and thus the current efficiency would be reduced as shown in Fig. 4b. The reduced efficiency can be related to the formation of gas bubbles at the anode surface, reducing the available reaction sites for chloride oxidation.

Whereas Bergmann and Koparal (2005) identified minimum chloride concentrations of about 7.5 mg/L to produce useful concentrations of FAC, Kraft *et al.* (1999b) used 20 mg/l in order to meet the chlorine demand in drinking water. In own lab studies up to 5 mg/L of FAC could be produced in synthetic water containing a chloride concentration of 20 mg/L at a current of 6 A (18.8 mA/cm²) (Fig. 4). Schmidt (2012) suggests a minimum chloride concentration of 20 mg/L to prevent the formation of chlorate, which is more likely to occur at low chloride conditions.

In the presented field studies (chapters 4-7), MOX cells with an active surface area of 639 cm² were used and a minimum chloride concentration of about 15 mg/L was identified as limiting factor for a useful system operation under real case conditions. Hereby, a residual FAC concentration of about 0.5 mg/L could be achieved with water containing no ammonium.

In low chloride regimes (e.g. chapter 4, [P2.1]) saturated NaCl solution was dosed into the source water to increase chlorine production. This, in principle, worked well and sufficient chlorine was produced.

3.2.3 Cell current, current density, and charge

Haaken (2015) and Kraft *et al.* (1999b) found a linear relation between current density (j) and chlorine production. Own lab studies conducted with the used MOX confirm this relation, when operated within operational limits shown in Fig. 5 with regards to current density and chloride concentration.

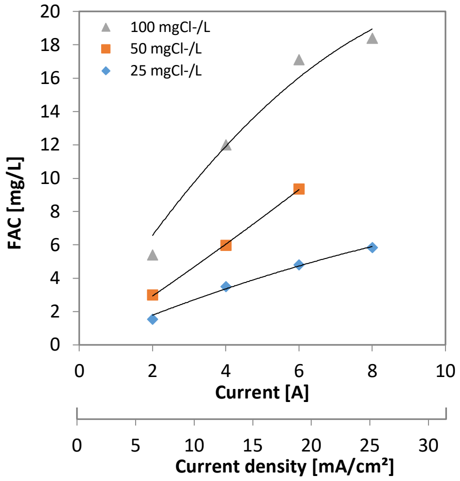


Fig. 5: Dependency of FAC concentration on the current and current density while applying different chloride concentrations

Fig. 5 shows the linear relation between current (current density) and the achieved FAC concentration for a current of up to 6 A. However, at currents of 8 A the chlorine production efficiency decreases due to gas bubble formation as already shown in chapter 3.2.2.

3.2.4 pH

Fig. 6 shows the dependence on pH and current applied on the chlorine production. The 20 % higher production rate of chlorine at pH 8 compared to pH 6 is caused by the lower potential required to produce chlorine at higher pH as shown in Fig. 7. While the pH has a strong effect on the disinfection capacity (Fig. 2), the effect of pH on the chlorine production has rather theoretical implications and did not play a major role during the application of ECl_2.

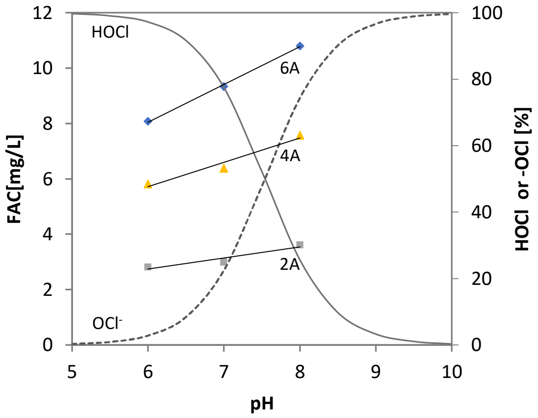


Fig. 6: Dependency of FAC on pH at different currents

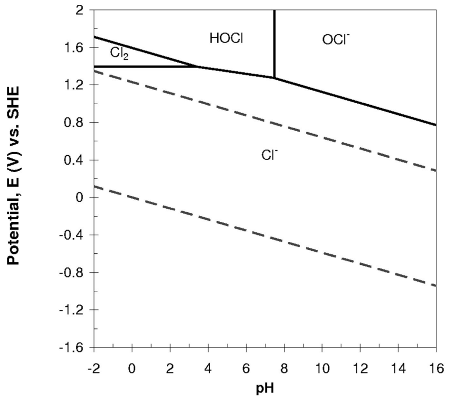


Fig. 7: Pourbaix diagram for chlorine in water

3.2.5 Flow rate and hydraulic retention time in the flow through reactor

With the flow rate the hydraulic retention time (HRT) inside the inline electrolysis reactor varies. Studies with the here applied MOX electrodes showed an inversely proportional linear relation between the flow rate and the chlorine concentration as shown in Fig. 8. Higher HRT (lower flow rate) as shown in Fig. 9 results in a drop of the chlorine production rate, which is related to the fact that available chloride is already converted.

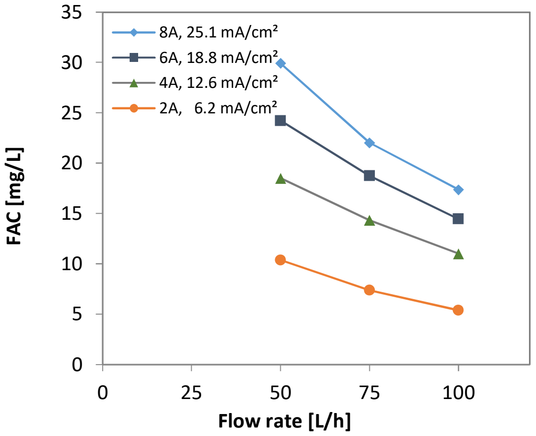


Fig. 8: Dependency of FAC concentration on flow rate at different currents applied

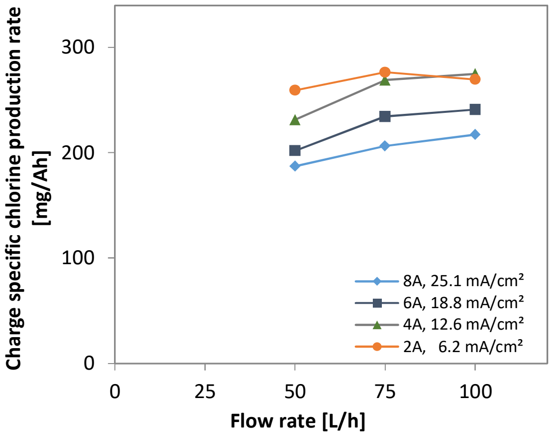


Fig. 9: Charge specific chlorine production rate in dependence of flow rate at different currents applied

3.2.6 Operational conditions and durability of electrolytic cells

During the here conducted field studies electrolysis reactors with eight monopolar IrO_2-RuO_2-Ti MOX electrodes, stacked at a distance of 5 mm, were used. The active surface area of each stack was 639 cm².

Due to the interdependency of many operational factors on the performance and durability of MOX cells, the manufacturers of MOX electrodes generally do neither publish nor guarantee lifetimes for the electrolytic cells. Following own experiments and data generated by others (Thürigen 2012; Wendt 2013; Haaken 2015) the current density and the duration of polarity inversion intervals have shown to have the highest influence on the durability of a MOX electrodes. Lifetime is hereby reduced at high current densities and shorter interval durations. Current densities on MOX electrodes between 20 mA/cm² (Wendt 2013) and ~100 mA/cm² (Ilian 2010) have been reported in literature. Haaken (2015) calculated 4,000 hours of operation of a Ti/RuO_2-IrO_2-TiO_2-MOX DSA with a current density of 20 mA/cm² and polarity inversion intervals every 60 minutes. With the here used MOX electrodes current densities of up to 25.2 mA/cm² (8A) were applied in lab studies and 18.8 mA/cm² (5A) in field test studies. Due to the lower current densities and longer polarity inversion intervals of 3 hours applied in field, the lifetime of the here used MOX cells are expected to be comparable or longer – as long as electrode scaling can be prevented (see chapter 3.3.1).

3.3 Operational challenges of ECl_2 systems

In the following the major drawbacks of ECl_2 operation are described.

3.3.1 Hardness and polarity inversion

A major problem in the practical use of electrochemical disinfection for water treatment is the formation of calcareous deposits on the cathode surface (Kraft *et al.* 1999a; Ilian 2010; Thürigen 2012; Wendt 2013). The formation of such is caused by a local increase in pH around the cathode due to the consumption of H^+ as shown in Eq. 3. If calcium, magnesium and bicarbonate ions are present in the water they will form calcium carbonate and magnesium carbonate deposits on the cathodes. Kraft *et al.* (1999b) summarized that higher water temperatures and higher flow velocities increase electrode scaling. As a consequence uneven current distribution with locally very high current densities will destroy the cell coating and finally result in a reduced chlorine production rate (Kraft *et al.* 1999a; Haaken 2015) and an increased energy consumption.

The application of polarity inversion at the cell helps to remove thin layers of deposits. Hereby the duration of an interval is adjusted to the hardness concentration and ranges between several minutes to a couple of hours. The hardness levels at which the long-term deployment of ECl_2 becomes infeasible is specific to the electrolytic cell applied and must be determined in long term field tests. Kraft *et al.* (1999b) showed that directly after the inversion of polarity the chlorine production rate is largely reduced, and stable chlorine production rates are reached only after a few minutes after polarity inversion. Therefore, the disinfection of the water cannot be assured for a period of a few minutes. The use of an intermediate storage tank is suggested to allow mixing of water containing chlorine and water that passed the cell shortly after polarity inversion. All ECl_2

stations developed and operated during the here studied pilot systems include such a tank.

Additionally, the inversion of polarity itself impacts the durability of the cell, and even a single inversion of polarity has resulted in a substantial drop of production rates at some MOX cell types (Thürigen 2012). This was caused by the dissolution of the catalytic active coating.

An alternative to the polarity inversion is the application of frequent rinsing with acid. However, due to the challenges in handling those acids in remote regions this approach was not considered here.

3.3.2 Formation of disinfection by-products

During the production of chlorine through electrolysis and the disinfection process DBPs are formed. Those are subdivided into inorganic DBPs like chlorate and perchlorate and organic by-products such as THMs or HAAs.

The formation of chlorate and perchlorate during chlorine electrolysis has been analyzed e.g. by Bergmann (2012), Czarnetzki and Janssen (1992) and Schmidt (2012). Hereby the chlorate concentration mainly correlates with the volume specific charge input (Qv) as available HOCl and OCl^- is further oxidized to chlorate (ClO_3^-) at the anode (Eq.6-7).

$$6\,HOCl + 3\,H_2O \rightarrow \;2\,ClO_3^- +\; 4Cl^- +\; 12\,H^+ +\; 1.5\,O_2 + 6e^- \qquad \text{(Eq. 6)}$$

$$6\,ClO^- + 3\,H_2O \rightarrow \;2\,ClO_3^- +\; 4Cl^- +\; 6\,H^+ +\; 1.5\,O_2 + 6e^- \qquad \text{(Eq. 7)}$$

Smaller current densities reduce the formation of chlorate and perchlorate on MOX electrodes. Bergmann (2010) reports that the formation of chlorate is negligible at current densities ≤ 200 mA/cm. As the here applied electrolytic cells were comparatively large, current densities did not exceed 25 mA/cm^2 and chlorate formation is not expected to occur in substantial concentrations.

Perchlorate is not directly formed at MOX electrodes (Schmidt 2012) and its formation depends mainly on the presence of chlorate. It is therefore not of concern during the operation of ECl_2 systems, which was confirmed in [P2.1] and by Haaken *et al.* (2012).

The very low chlorate and perchlorate formation potential with MOX electrodes is a major advantage towards e.g. BDD electrodes, which were found to produce substantial amounts of chlorate and perchlorate (Bergmann *et al.* 2010; Haaken 2015)

The formation of organic DBPs correlates with the presence of natural organic matter (NOM) and the nature of the molecules with respect to its reactivity with chlorine and applied chlorine concentrations (Bond *et al.* 2011; Bond *et al.* 2014). To prevent higher concentrations of organic DBPs the concentration of NOM in the source water should be monitored and eventually reduced. Studies have shown that the THM concentration is likely to exceed 100 µg/L in waters that contain a TOC of > 4 mg/L if the water is distributed over a period of 2-3 days (Ratnayaka *et al.* 2009).

Selected guideline values of the applied chlorine concentrations and the here mentioned DBPs are shown in Tab. 3.

Tab. 3: Selected guideline values for DBPs in drinking water

Parameters	**Germany (BMJV 2001)**	**EU (EC 2020)**	**WHO (WHO 2017)**	**India IS 10500 (BIS 2012)**
Chlorine (FAC) [mg/L]	1.2[a]/0.1-0.3[b]		≥0.5 [d]	0.2/1.0[e]
Bromate [µg/L]	10	10	10	-
Chlorate [µg/L]	70	250	700	-
Chlorite [µg/L]	200	250	700	-
THM [µg/L]	10 [b]/50[c]	100	60-300	-
Bromoform [µg/L]				100
Dibromochloromethane [µg/L]				100
Bromodichloromethane [µg/L]				60
Chloroform [µg/L]				200
HAA				

[a]during treatment, [b]at the end of treatment, [c]point of use, [d]when distributed, [e]permissible limit in absence of alternate source

As the formation of organic DBPs is not an intrinsic effect of the ECl_2 process itself but rather related to the application of chlorine in general, organic DBPs were, except for some sporadic measurements of THM concentrations during the field tests, not systematically considered in this dissertation work. Sufficient literature on this topic is already available, e.g. in Li and Mitch (2018), Thompson *et al.* (2015), or Prasad (2020).

4 Combination of riverbank filtration with ECl_2 for river bound communities

4.1 Concept

Surface water bodies are continuously subject to pathogenic contamination through the inputs from pastures, fecal matter from livestock, wildlife and the discharge of treated and untreated WW (Pandey *et al.* 2014). Especially in developing regions, where 144 million people rely on surface water to cover drinking water demands (WHO 2019), river bound communities are constantly at risk to suffer from waterborne diseases. Whenever site conditions permit, the filtration of surface water through the adjacent riverbank may remove most of the pathogens. During RBF, water flows from the river through the riverbank/riverbed into a pumping well.

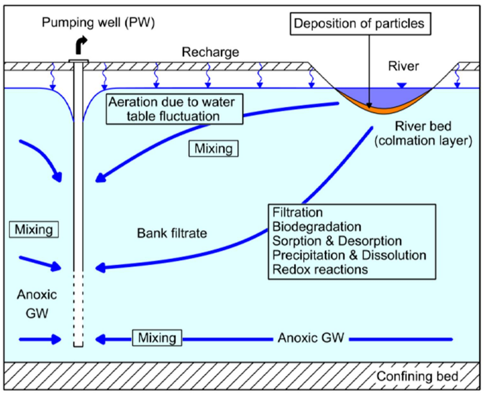


Fig. 10: Processes affecting water quality during passage through the riverbed and aquifer sediment (Grischek and Paufler 2017)

Major treatment of the water occurs hereby during the passage through the highly biologically active colmation zone and the riverbed (Fig. 11). Already, within the first centimeters of this layer a substantial reduction of particles, degradation of organics and sorption processes occur. In the subsequent aquifer further degradation, sorption, and desorption processes as well as mixing with groundwater take place. In most parts of the world RBF wells are designed for particle and pathogen removal only. Therefore, they are placed in rather short distances to the water body with short HRTs. In India, the application of RBF dates back to the 1950ies, where it has become an alternative to surface water abstraction (Sandhu and Grischek 2012) and several large scale RBF abstraction sites have been implemented, e.g. in Haridwar (22 collection wells at River Ganga), Nainital (9 vertical filter wells (VFW) Lake Nainital), Delhi (90 VFW Yamuna) and Ahmedabad (7 radial collector wells Sabarmati).

Considering the water supply situations in rural India the application of RBF may be beneficial for river bound communities. Even though in many regions it may provide pre-filtration only, it reduces otherwise required investments into filtration technology and infrastructures (Sandhu *et al.* 2011). Studies have shown pathogen indicator removal rates of up to 5.5-6.1 log for total coliforms and for > 4.4 log for bacteriophages (Weiss *et al.* 2005; Sandhu *et al.* 2011) and by that substantially improved source water conditions. Despite very high removal rates, monitoring campaigns and risk assessment studies have repeatedly shown the presence of small quantities of total coliforms and *E. coli* in RBF wells even at greater distances (48-190 m) to the riverbank (Sandhu *et al.* 2011; Bartak *et al.* 2015). Therefore, additional disinfection measures are required. Chlorine, in contrast to e.g. UV treatment or ultrafiltration (UF), has a long proven record providing pathogen free water also during drinking water storage and distribution (Clasen *et al.* 2007; Hashmi *et al.* 2009). However, due to the above-mentioned challenges of residual disinfectant supply and adequate dosing in rural developing communities, alternatives to “simple” chlorine dosing are required.

4.2 Treatment approach

The inline electrolytic production of chlorine (ECl_2) directly from the natural chloride content of the bank filtrate could pose a robust and reliable alternative to the supply and dosing of chlorine. If electricity can be supplied through solar PV battery systems, pumping and treatment could be conducted completely autonomously and operation e.g., in areas that are difficult to access becomes possible. The objective of the presented case study [P2.1] was to combine for the first time an existing RBF setting with an ECl_2 system to merge the benefits of both technologies. The combination was supposed to assure a reliable and robust drinking water supply for river bound communities even under the challenging monsoon conditions. The setting hereby was supposed to assure water free of pathogen indicators and chlorine concentrations (Tab. 3).

To evaluate the applicability of the RBF-ECl_2 system combination, a long-term field test was conducted in Haridwar, India, over a duration of 17 months. Due to the low natural chloride concentration of 14 ± 2 mg/L (n=20) in the bank filtrate, the dosing of NaCl solution prior to the inline electrolytic chlorine production was tested for the first time under field conditions. Saturated NaCl solution was dosed into the feed tank (Fig. 11) whenever a predefined ratio between voltage and current was surpassed (e.g. V=9V, I=3A, ratio=3) and dosing stopped again once the ratio was reached. From here the water passed the filtration and the electrolytic unit. On average a chloride concentration of 30 mg/L was anticipated to allow for sufficiently high drinking water disinfection capacities with the applied electrolytic cell. This required on average the addition of 26 g of

NaCl per m³ water treated. Chlorine production was adjusted through online reading of the ORP in the final storage water tank. An AFM® filtration unit was installed prior to the disinfection to further reduce turbidity of the bank filtrate.

The setting was installed in Haridwar and served as a base for intensive sampling campaigns. The evaluation of two operation periods resulted in publication [P2.1]. The bank filtrate for ECl_2 disinfection was abstracted from a large diameter caisson well (IW #18) on the Pant Dweep Island located between the Upper Ganga Canal and the Ganga River. The distance to the nearest canal bank is 115 m. The position of the well on an island and the significant natural gradient of the water table results in a high portion of bank filtrate in the abstracted water (Sandhu and Grischek 2012). The publication [P2.1] describes the results achieved during an 8-month intensive sampling campaign and a shorter 2-week sampling campaign conducted after system optimization.

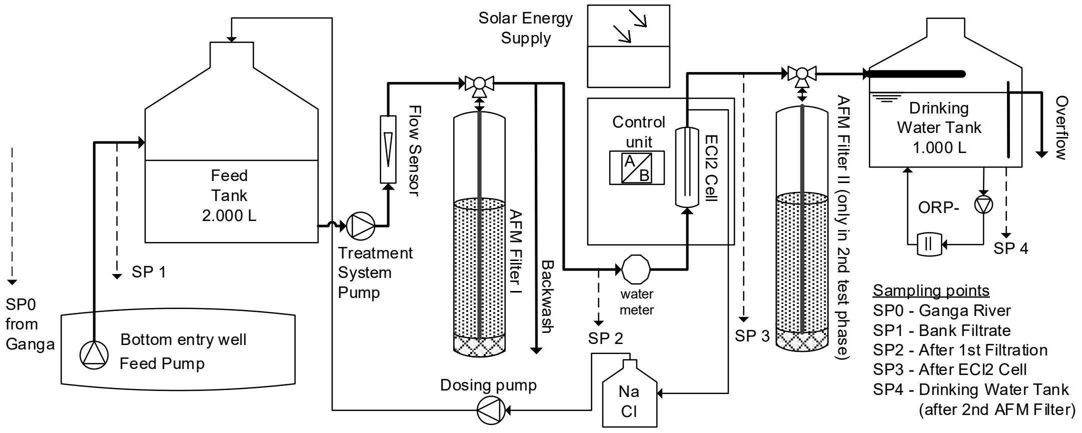


Fig. 11. Pilot system setting in Haridwar

4.3 Results

The results from the first sampling campaign confirm that RBF is capable to serve as a very efficient pre-treatment step substantially reducing pathogens and turbidity and equalizing fluctuations in source water characteristics (Tab. 4).

Tab. 4: Source water qualities and operational parameter of pilot station in Haridwar

Parameter **Pathogen Indicator**	**Ganga River SP0 (n)**	**Bank Filtrate SP1 (n)**
Total Coliforms [MPN/100 mL]	1.07x10^{6} ± 1.89x106 (10)	8.87x10^{1} ± 1.20x10^{2} (30)
E. coli [MPN/100 mL]	2.34x10^{4} ± 6.34x104 (13)	1.09 x10^{1}± 1.79x10^{1} (26)
Chemical parameters		
Hardness [mg/L]	92 ± 39 (19)	207 ± 40 (41)
Chloride* (titrated) [mg/L]	7 ± 3 (15)	14 ± 2 (20)
NH4-N [mg/L]	0.15 ± 0.07 (34)	0.23 ± 0.18 (41)
Physicochemical parameters		
EC [µS/cm]	156 ± 17 (15)	403 ± 31 (38)
pH	7.8 ± 0.3 (23)	7.5 ± 0.1 (43)
Temperature [°C]	25.5 ± 2.6 (21)	24.8 ± 1.1 (41)
ORP [mV]	ND	476 ± 58 (38)
UVA-254 [1/m]	51.6 ± 28.1 (14)	0.8 ± 0.9 (15)
TOC [mg/L]	ND	1.96 ± 0.49** (16)
Operational parameters		
Flow rate [L/h]	100-250	

* Mixed values, MPN-most probable number, ND-not determined

Especially the effects of heavy rainfall events during monsoon period were completely equalized through RBF and turbidity was substantially reduced from 501 ± 243 NTU in the river water to 0.6 ± 0.6 NTU in the bank filtrate (Fig. 12). The AFM filter reduced the already low turbidity values from the bank filtrate further to 0.3 ± 0.3 NTU. The increase in turbidity in the drinking water was related to calcareous deposits released during polarity inversion. In the second evaluation period (not presented here) an additional AFM filter (Fig. 11) was successfully installed to remove such deposits.

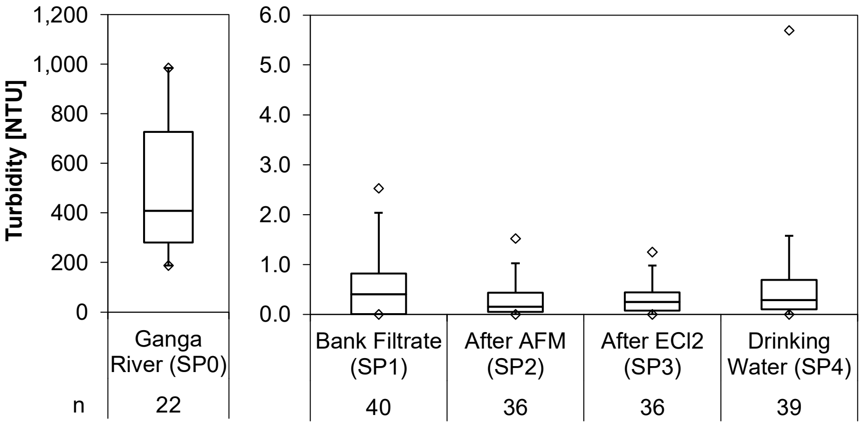


Fig. 12: Turbidity during treatment with ECl$_2$ pilot station

RBF achieved a log reduction of 3.9 and 3.6 for total coliforms and *E. coli*, respectively. The ECl$_2$ system completely removed present fecal indicators and the water could be always kept microbially safe. The maximum log reduction of the RBF-ECl$_2$ constellation was > 5.0 for total coliforms and > 3.5 for *E. coli* (Fig. 13).

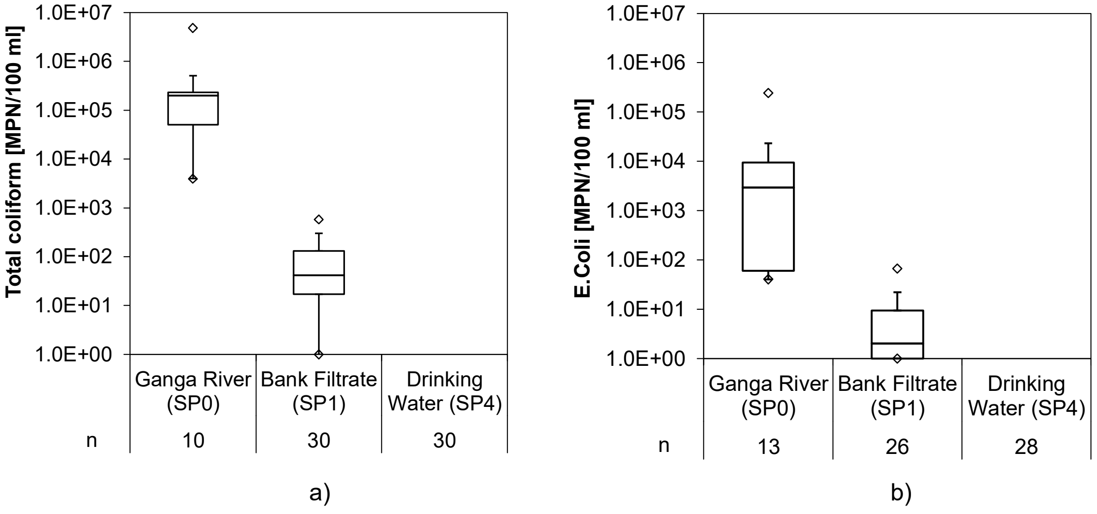


Fig. 13 a) and b): Total Coliform (a) and *E. coli* (b) numbers found in Ganga River, after RBF and ECl_2 (drinking water)

FAC and total chlorine concentration reached 0.27 ± 0.17 mg/L and 0.30 ± 0.16 mg/L, with maximum concentrations of 1.01 mg/L (Fig. 14).

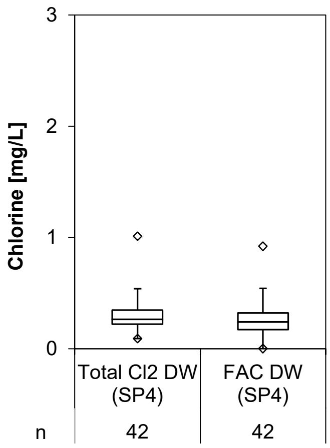


Fig. 14: Total chlorine and FAC Concentrations achieved in ECl_2 treated water (drinking water)

The small difference between total chlorine and FAC hereby indicates only minor presence of ammonium in the source water.

The pilot system was also tested for its potential to form inorganic DBPs. With chlorate concentrations of 22 ± 29 µg/L (n=19) the inorganic DBP formation potential is not of concern (Tab. 3). Organic DBPs have also not been of concern mainly due to the low dissolved organic carbon (DOC) in the bank filtrate [P2.1].

The pilot station in Haridwar needed frequent manual cleaning. Mainly hardness deposits covered the cell and polarity inversion did not completely remove the deposits. Additionally, the deposits that were released after polarity inversion caused clogging issues within the treatment system itself and increased the turbidity of the treated water.

Further operational challenges were caused by the dosing of salt and the subsequently increased production of chlorine. This increase was not quickly enough sensed by the ORP sensor in the storage tank, for which the station continued to dose NaCl and to produce more chlorine. This resulted in intermittently high chlorine concentrations above the maximum permitted value and caused a substantially higher NaCl demand than originally anticipated [P2.1]. In consequence the chlorine and chloride values largely fluctuated around the target values.

4.4 Conclusion

The major findings of the pilot trial in Haridwar can be summarized as follows:

- RBF largely reduces raw water quality fluctuations and simplifies disinfection through ECl_2 compared to the direct treatment of surface water.
- With the combination of RBF and ECl_2 pathogen indicators can be completely removed and the treated water can be safely supplied as disinfected drinking water.
- Most of the time chlorine concentration could be maintained within the required limits. Reasons for larger fluctuations were identified. Options for optimization were developed and successfully tested during a second short sampling campaign [P2.1].
- AFM filtration is useful to further reduce turbidity at marginal extra costs.
- The application of an ORP allows an indication of insufficient disinfectant supply given the sensor has sufficient time to react to given changes. For more accurate and faster readings the application of chlorine probes is more appropriate.
- Organic DBP formation was of no concern in the given settings, mainly due to the low DOC concentration in the bank filtrate.
- The here for the first time applied and evaluated dosing of NaCl solution worked in principle, but further optimization was required to reduce the NaCl demand to anticipated levels.

The combination of RBF and ECl_2 is well suited for the treatment of surface water for river bound rural communities as long as the electrode scaling can be prevented.

4.5 Publication related to this topic

[P2.1]

Otter, P., Malakar, P., Sandhu, C., Grischek, T., Sharma, S., Kimothi, P., Nüske, G., Wagner, M., Goldmaier, A. and Benz, F. **2019** Combination of River Bank Filtration and Solar-driven Electro-Chlorination Assuring Safe Drinking Water Supply for River Bound Communities in India. *Water*, **11**(1), 122.

5 Removal of arsenic from contaminated groundwater sources

5.1 Contaminated water sources and present arsenic mitigation strategies

Arsenic is a naturally occurring element and present in food, soil, air, and water. It is known to be highly toxic. Humans are exposed to different arsenic species, of which the dissolved inorganic forms in drinking water are most significant for natural exposure and are the main cause for adverse effects on human health. The exposure to arsenic through the consumption of groundwater is a problem especially in South Asia (Mazumder *et al.* 2010), South-East Asia (Berg *et al.* 2001; Hug *et al.* 2008; Jiang *et al.* 2012) and Latin America (Astolfi *et al.* 1981; Del Razo *et al.* 1990). The worldwide mostly affected region is the Ganga-Meghna-Brahamaputra plain in Bangladesh and West Bengal (WB), India. In Bangladesh three out of five million wells drilled into Ganga alluvial deposits, which are used for public water supply, are assumed to be contaminated by arsenic released from the aquifer sediments. Up to 70 million people in Bangladesh (Lepkoski 1998; Ratnaike 2003) and 42 million people in West Bengal are affected (Ratnaike 2003). The inhabitants of rural areas are mostly prone to consume contaminated water as they rely on shallow tube wells (WHO 2012). The WHO guideline value for arsenic was reduced in 1993 from 50 to 10 µg/L and is considered as provisional. Whereas most countries have taken the WHO guideline value into their legal frameworks, in Bangladesh a total arsenic concentration of ≤ 50 µg/L is still permissible. In India this guideline value is acceptable “if no other sources is available” (DPHE 2009; BIS 2012).

Various arsenic mitigation technologies have been studied on lab as well as field scale and are widely reported and summarized in literature (Jiang 2001; Amrose *et al.* 2015). Systematic reviews on the field efficiency of research projects conducted between 1980 and 2011 showed that oxidation-filtration and ion exchange technologies showed poor effectiveness i.e., samples of treated water contained more than 50 µg/L of arsenic. Coagulation and co-precipitation-filtration approaches performed slightly better with about half the technologies showing good results with more than 95 % of the samples arsenic concentrations of ≤ 50 µg/L.

When dissolved iron is available in the groundwater the pre-oxidation and coagulation-precipitation of iron and arsenic is still considered to be the most cost effective technological approach for the mentioned regions (Lytle *et al.* 2005; Jain and Singh 2012). Through this process arsenic attaches to the surface of Fe(III) and is entrapped within growing Fe(III) particles by inclusion, adsorption and occlusion (Lytle *et al.* 2005), through which arsenic becomes filterable. Hereby, in situ formed Fe(III) particles have shown to remove substantially higher amounts of arsenic compared to pre-formed Fe(III) particles (Edwards 1994).

Whereas oxygen can readily oxidize Fe(II) to Fe(III) it does not effectively oxidize As(III) to As(V). Still, this oxidation is favored as As(V) sorbs in higher quantities to Fe(III) particles (Edwards 1994; Hering *et al.* 1997; Lytle *et al.* 2005; Cao *et al.* 2008; Ravenscroft *et al.* 2009). Chlorine as strong oxidizing agent is readily oxidizing As(III) to As(V). Monochloramine and chlorine dioxide, however, have shown to be ineffective in doing so (Ghurye and Clifford 2001; Dodd *et al.* 2006). Studies have further shown that iron particles formed during the oxidation with chlorine had a by 40 % higher capacity to remove As(V), than particles formed through oxidation with oxygen alone (Lytle *et al.* 2005).

5.2 Suggested treatment approach

The above-mentioned effects of chlorine during co-precipitation may enhance overall arsenic removal efficiency. As the supply and application of chlorine in rural areas is challenging, chlorine induced co-precipitation approach have not been successful in the past. To overcome this challenge the application of ECl_2 is seen as a promising and innovative alternative to the supply of the otherwise required chemicals.

In the presented publications autonomous, solar driven systems combining ECl_2, precipitation and coagulation were evaluated for its arsenic removal performance treating different source waters. The settings were tested at three pilot sites in Niederfraundorf (NFD), near Dresden, Germany [P1.1], in San Jose, Costa Rica [P1.2] (Fig. 15b), and in West Bengal, India [P1.3] (Fig. 15a). In all settings naturally present Fe(II) was oxidized through chlorine, which was produced via ECl_2 followed by precipitation and coagulation. The precipitate, together with co-precipitated arsenic, was filtered through a manganese greensand plus (GSP) based filtration system. The GSP acted as intermediate redox buffer in periods of insufficient chlorine production e.g., during polarity inversion of the electrolysis cells. A continuous monitoring of the ORP value in the final water storage tank hereby indicated the supply of sufficient oxidizing agent to the treatment process and was used as parameter to adjust chlorine production.

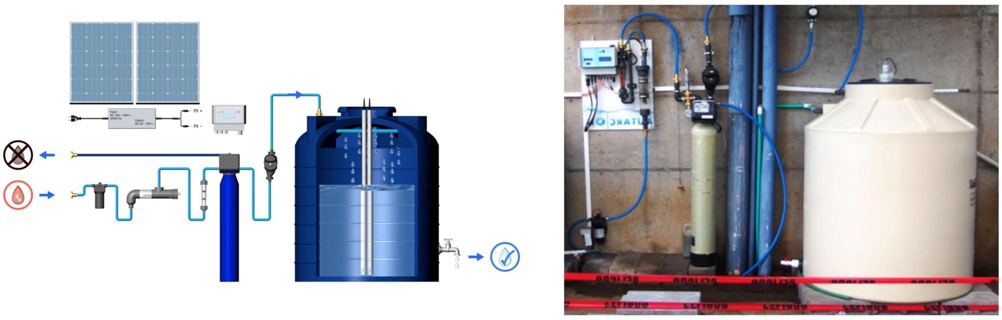


Fig. 15: Pilot test setting as installed in West Bengal (a) and Costa Rica (b) during the in [P1.2 and P1.3] presented study

5.3 Performance evaluation and results

During a first pilot test campaign in NFD [P1.1] with a premature treatment system, total arsenic could be reduced by 96 % from 300 ± 270 µg/L to 11 ± 4 µg/L. At the same time total iron was removed by over 99 % from 7.1 ± 7.0 to 0.04 ± 0.04 mg/L. The raw water data show that iron and arsenic concentrations were largely fluctuating but the setting proved to supply stable effluent concentrations. Despite the low natural chloride concentrations of only 8.4 ± 0.8 mg/L sufficient chlorine could be produced to maintain a chlorine residual. This however required a low flow rate through the ECl_2 reactor of 50 L/h only.

In Costa Rica only 68 % of the arsenic was removed. This was related to the comparably low Arsenic inlet concentrations of 40.1 ± 23.0 µg/L and a low average Fe/As mass ratio of 14.3. Although the strict WHO limits for arsenic could not be continuously maintained throughout, the achieved results were promising and led to the installation of a comparable pilot setting in Nadia District, WB, India, described in [P1.3]. This region is one of the worldwide most severely arsenic effected regions (Bhakta *et al.* 2016). The implementation at this site allowed a more intensive evaluation of the arsenic removal efficiencies and operability in a real case scenario under challenging groundwater conditions (Tab. 5). The field test in WB comprised four different testing periods (P1 to P4), that were conducted consecutively over a total duration of 10 months from September 2016 to July 2017.

Tab. 5: Groundwater quality from test well during test period in WB 2016-2017 and reference values from the literature

Parameters	Minimum/Maximum	Mean ± SD	n	Literature Values**
As_{total} [µg/L]	65/330	187 ± 45	50	62 ± 127
Fe_{total} [mg/L]	2.0/7.6	5.5 ± 0.8	51	3.7 ± 5.4
Mn_{total} [mg/ L]	0.4/3.0	1.5 ± 0.4	41	0.6 ± 0.8
NH_4-N mg/ L]	0.7/2.9	1.4 ± 0.5	47	2.0 ± 1.0
Hardness [mg/L $CaCO_3$]	285/495	381 ± 54	32	-
O_2* [mg/L]	1.8/4.5	2.7 ± 0.7	53	-
pH	6.2/7.2	6.8 ± 0.2	53	7.0 ± 0.2

* Oxygen intrusion through sampling likely

** Average values taken from Hug *et al.* (2008) for wells (10-90 m) in Bangladesh (BGS-DPHE database)

Due to the high concentration of total hardness and ammonium in the source water the conduction of the pilot trial was challenging. Ammonium exhausted the chlorine production capacity of the ECl_2 system and FAC was converted to monochloramine. Throughout all test periods, the breakpoint could only be reached during sampling period 2. Elevated hardness levels quickly covered the electrolytic cells, which critically reduced the chlorine production capacity and substantially increased the maintenance efforts. Additionally to that, frequent filter breakthroughs occurred in periods 1-3 through which iron (Fig. 17) and arsenic were released into the final water storage tank. Excerpts of the results achieved by describing the inlet and outlet concentrations of arsenic and iron are given in Fig. 16 and Fig. 17.

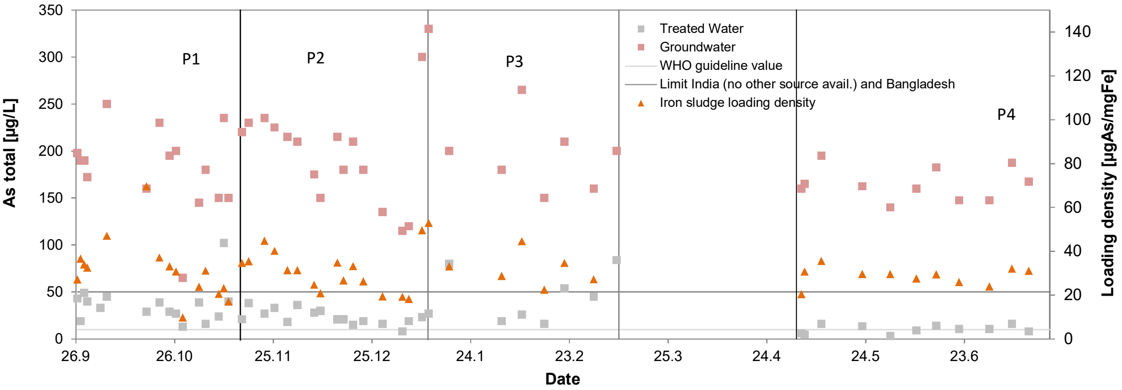


Fig. 16: Arsenic removal performance during field test in WB [P1.3]

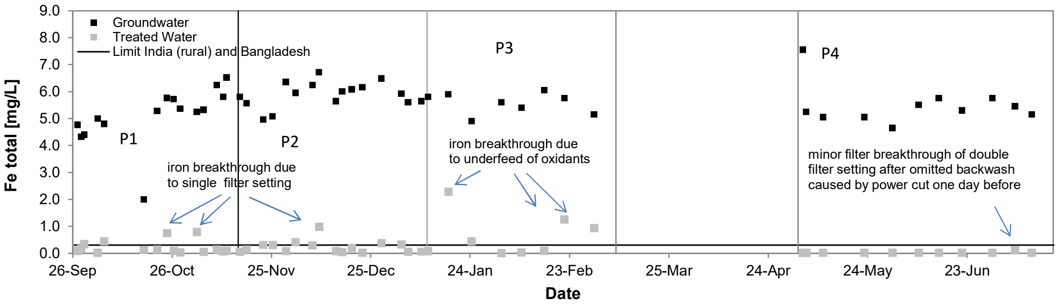


Fig. 17: Iron removal performance during field test in WB [P1.3]

After optimizing the system setting by increasing the current at the electrolytic cell and adding a second GSP filter in period 4, arsenic and iron removal rates reached 94 % and >99 % respectively. Even though the total arsenic outlet concentrations of 10±4 µg/L the in India valid maximum permissible levels of 50 µg/L in rural areas were met, it was not possible to continuously achieve the WHO guideline values of 10 µg/L. Additionally, phosphate competed with arsenic for adsorption sites on the formed iron precipitate and a minimum Fe/As mass-ratio of ≥30:1 was required to allow for efficient arsenic removal.

The trials also showed that arsenic removal was reduced when oxidant supply was insufficient. This was also shown during the field test in NFD when arsenic was released from the setting during short-term malfunctions of the electrolytic cell. This proved the positive effect of chlorine on overall arsenic removal in a co-precipitation setting as described by Lytle *et al.* (2005).

5.4 Conclusion and prospects

The performance of the tested settings was encouraging at the presented pilot sites considering the challenging field conditions. However, the maintenance efforts that were required to keep the pilot station in WB running would not allow a sustainable long-term operation. Due to the increased levels of hardness polarity inversion alone was not sufficient to prevent the formation of calcareous deposits and frequent manual cleaning was required. Ammonium prevented the supply of free residual chlorine through which the control of chlorine production through ORP measurements failed. Therefore, future research should focus on the evaluation and development of

1. a preceding ammonium removal step to reduce the chlorine demand of the water and
2. the application of OCG to prevent scaling as described in chapter 8.

The removal of ammonium is commonly based on a nitrification step which requires aeration. Through this process most of the Fe(II) would be oxidized and must be removed by subsequent filtration. Nevertheless, as arsenic will be oxidized only partially not all of it can be co-precipitated and retained in the filters. Here adsorption technologies may be the only option to effectively remove still present arsenic if the given legal requirements cannot be met.

The formation of calcareous deposits and the reduction of nitrate back to ammonium at the cathode during ECl_2 (Li *et al.* 2009; Duca and Koper 2012) further limits the application of ECl_2 when treating waters as found in WB . This provides an additional argument for the application of OCG as described in chapter 8. Additionally to these developments the formation of organic DBPs during chlorination must be evaluated and prevented as much as possible.

5.5 Publications related to this topic

[P1.1]

Feistel, U., **Otter, P.**, Kunz, S., Grischek, T. and Feller, J. **2016** Field tests of a small pilot plant for the removal of arsenic in groundwater using coagulation and filtering. *Journal of Water Process Engineering*, **14**, 77–85.

[P1.2]

Kunz, S., Romero-Esquivel, L. G., **Otter, P.**, Feistel, U., Grischek, T., Valverde-Cerdas, J. and Feller, J. **2018** Treatment of arsenic-contaminated water using in-line electrolysis, co-precipitation and filtration in Costa Rica. *Water Sci Technol Water Supply*, **18**(1), 40–48.

[P1.3]

Otter, P., Malakar, P., Jana, B., Grischek, T., Benz, F., Goldmaier, A., Feistel, U., Jana, J., Lahiri, S. and Alvarez, J. **2017** Arsenic Removal from Groundwater by Solar Driven Inline-Electrolytic Induced Co-Precipitation and Filtration—A Long Term Field Test Conducted in West Bengal. *International Journal of Environmental Research and Public Health (IJERPH)*, **14**(10), 1167.

6 OCG for decentralized disinfection of treated wastewater to allow its reuse

6.1 Concept

Growing population, urbanization, socio-economic development, climate change and degrading water qualities increase the stress on high quality water resources for drinking purposes (Mudgal *et al.* 2015; Delacamera *et al.* 2016). In 17 countries, comprising 25 % of the world population, "extremely high" levels of water stress are challenging future development. Here, agriculture, industries and municipalities consume more than 80 % of the average annual water supply, leaving those countries very vulnerable towards fluctuations caused e.g. by droughts and/or increased water withdrawals (Hofste *et al.* 2019). At the same time, more than 80 % of WW from human activities is discharged without any treatment. Consequently, between 1.8 and 2 billion people globally depend on drinking water sources that are fecally contaminated. The absence of adequate WW treatment is usually significantly higher in rural communities (World Health Organization & 17 UN-Water 2014). Even in Spain 3 to 4 million inhabitants in more than 6,000 agglomerations live in small communities without adequate WW treatment. Many of such comprise less than 500 person equivalents (PE) and are located in water scarce areas. In such communities often the locally available freshwater supplies are already over abstracted (Asano 2007) and WW reuse poses an attractive alternative to reduce fresh water needs. With an average reuse of 0.5 % of the total freshwater extracted the reuse sector in the EU is still underdeveloped (Mudgal *et al.* 2015). Spain already reuses with 413 Mm^3/a about 10 % of the countries WW volume and by that 50 % of the total reclaimed WW in the EU (Jeffrey and Raffin 2018). The water is mainly reused for irrigation of agricultural sites, irrigation of golf courses, groundwater recharge and river flow augmentation (Mudgal *et al.* 2015; Paranychianakis *et al.* 2015).

Existing reuse schemes and technologies are currently limited to the application along with conventional centralized WW treatment plants, despite the fact that case studies have shown higher reuse potential in decentralized applications (Suriyachan *et al.* 2012). Therefore decentralized solutions have become of interest as they eliminate long-distance water transportation and can be installed close to the source of WW and/or the point of reuse (Lavrnić and Mancini 2016). That way, they can also be easier adapted to local WW conditions and reuse requirements (Prieto *et al.* 2013; Capodaglio 2017). In that context natural treatment solutions such as CWs can be a cost-efficient alternative for treatment of WW in rural areas (Kadlec & Wallace, 2009, Vymazal, J. 2005). The major advantage of CWs, towards alternative decentralized solutions such as membrane bioreactors (MBR), is their robust and reliable operation, with minimal energy and maintenance requirements and their capacity to balance peak flows and peak loads (Masi *et al.* 2007).

Even though there is no common set of regulated water quality parameters in existing reuse guidelines, all limit the number of pathogen indicators, especially if the water is intended for reuse applications e.g., in sanitary, cooling, indirect potable use or the irrigation of public accessible areas (Stevo & Maurozio 2015). Although pathogen removal rates of around 99 % (two log units) have been reported for CWs (Alexandros and Akratos 2016) they are not capable in reliably meeting *E. coli* numbers ≤ 10 or even 0, which is commonly required for the mentioned reuse applications. To allow safe water reuse and to minimize public health risks associated with potential exposure to treated WW an additional disinfection step is required (Tchobanoglous 2003; Capodaglio 2017; Mishra *et al.* 2018). Otherwise, established disinfection technologies such as membrane filtration, UV radiation, and ozonation cannot assure residual disinfection during the distribution of the reclaimed WW (Antonelli *et al.* 2008; Haaken *et al.* 2013). Therefore, many water reuse guidelines require chlorination. In rural regions the supply of such has proven to be challenging, which limits the reuse potential of otherwise adequately treated WW.

6.2 Treatment approach

The production of chlorine from either the natural chloride content of the water itself or added NaCl can hereby substitute the external supply of chemicals. Compared to the disinfection of drinking water, substantial higher amounts of chlorine are required due to the additional chlorine demand mainly exerted by ammonium and organic substances present in wastewater treatment plant (WWTP) effluent. A polishing step implemented between the WW treatment process and the chlorination may hereby largely reduce the chlorine demand.

In a pilot study [P3.3] the treatment performance of a vertical flow constructed wetland (VFCW) was combined with a solar driven chlorine-based disinfection system on a central WWTP in Andalusia, Spain. The VFCW was evaluated for its buffering capacity and its ability to polish treated or partly treated WWTP effluent. Different to the other presented studies [P3.1 and P3.2], where horizontal flow constructed wetlands (HFCW) were tested, the VFCW is expected to fully nitrify present ammonium allowing the disinfection with free available chlorine.

With total hardness of 694 ± 54 mg/L $CaCO_3$ in the treated WW challenging operation conditions for ECl_2 systems were expected. Even lower concentrations have already been identified to be problematic during ECl_2 of treated WW in own studies [P3.1 and P3.3] and elsewhere (Ilian 2010; Haaken 2015). To reduce the formation for deposits a first derivation from an ECl_2 system was implemented and tested during this field test. In the given setting a side stream of only 4-23 % of the total water volume treated was fed into an intermediate storage tank. From here it was continuously circulated through the electrolytic cell to produce chlorine until an equilibrium chlorine concentration was established. The elevated "natural" chloride levels in the treated WW of up to 819 ± 132 mg/L were expected to be sufficient for producing appropriate chlorine concentrations for efficient disinfection. Therefore, chlorine was not anymore produced by ECl_2 but through the so-called OCG. With this approach the amount of deposit forming precursors passing the cell was substantially reduced. Additionally, it was foreseen to soften this smaller portion of the water.

Between the here implemented system combination of VFCW and OCG an additional AFM® filter was installed to reduce particles eventually released from the VFCW. The complete setting (Fig. 18) was powered by a small solar PV battery system.

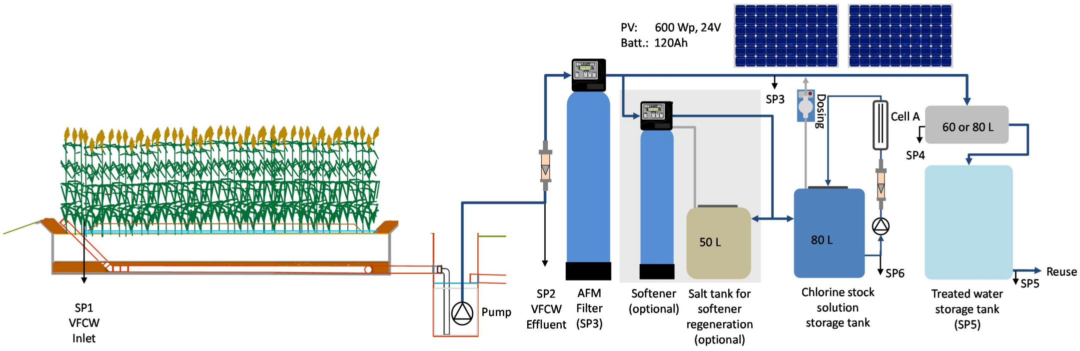


Fig. 18: Combination of VFCW and solar driven OCG system setting tested in Spain

6.3 Performance evaluation and results

During the pilot trials, different chlorine dosing rates and flow rates were tested to understand its effect on the disinfection capacity of the system. The major findings of the system performance can be summarized as follows:

- The VFCW was capable in reducing even elevated concentrations of TSS by 92 %. The additional filtration process through AFM® media further reduced the TSS to a total reduction of 99 %. It was found to be particularly useful considering minor additional costs and durability of the filter.

- In the VFCW the COD could be reduced to below 20 mg/L independent from the inlet concentrations, which ranged from 25±6.5 to 87±22 mg/L during the evaluated trial periods.
- In the VFCW ammonium could be reduced by 99.5 % to reach effluent concentrations of 0.17±0.12 mg/L-N. The chlorine demand exerted after nitrification could therefore be reduced substantially and allows the disinfection with FAC.
- During the optimum treatment setting less than 9 mg/L of chlorine was sufficient to safely disinfect the treated WW. This is a substantial reduction compared to the otherwise at this site practiced addition of 40 mg/L of FAC.
- All evaluated pathogen indicators could be maintained below the guideline values for the optimum treatment settings. The VFCW achieved hereby a log unit removal of up to 2.3 and 3.2 for total coliforms and *E. coli*, respectively. With OCG total log unit removal reached 5.1 and ≥4.6 for total coliforms and *E. coli*, respectively (Fig. 19).

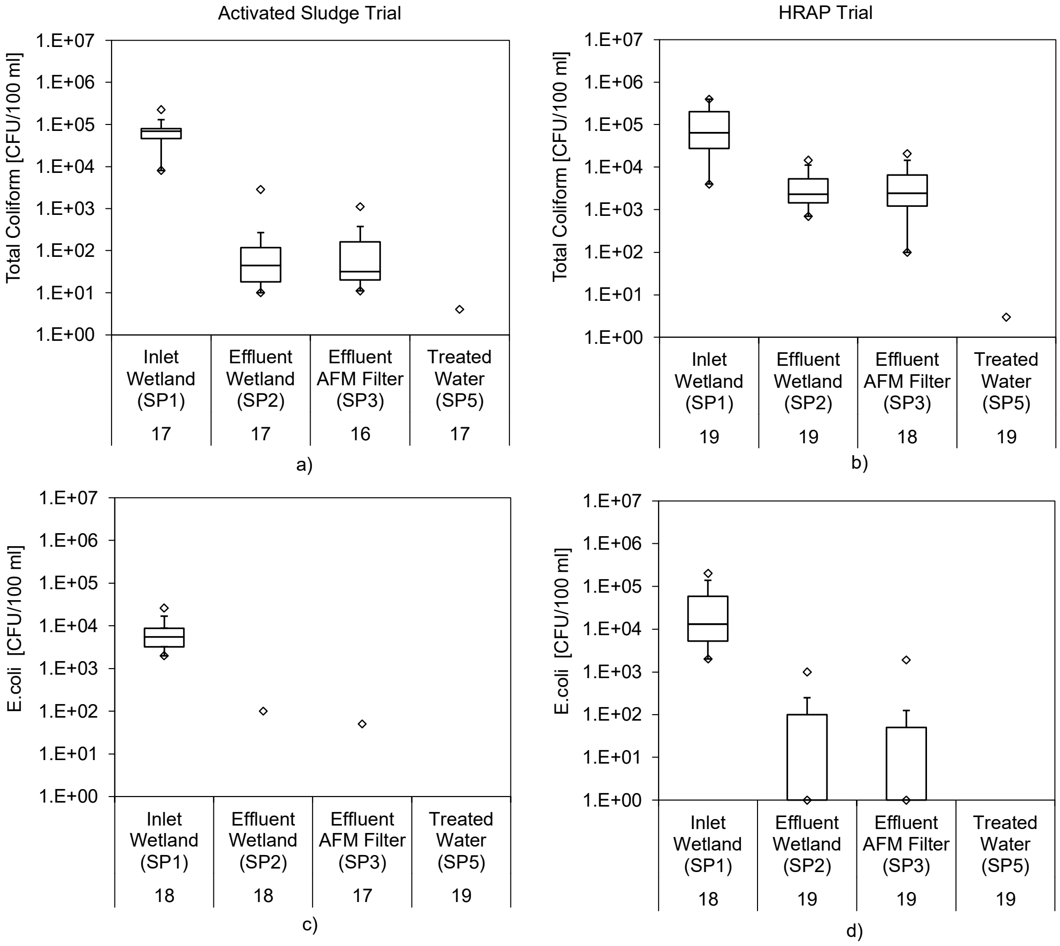


Fig. 19: Total coliform and *E. coli* counts determined along the treatment path

- The electrical conductivity in the wetland increased beyond the permissible limits after fully establishing the vegetation due to excessive evapotranspiration (ET) in the VFCW.
- Neither salt for chlorine production nor for softener regeneration was required. The treatment system can be operated without any chemical input.
- Sufficient electricity was supplied by a 24 V, 600 Wp, 120 Ah solar PV battery system to run the system completely independent from external energy supply at the pilot site.
- Scaling of electrodes did not occur with the selected setting.
- Except for the electric conductivity and high chloride concentrations the achieved water quality allows reuse applications, which often go beyond the requirements of simple irrigation, such as indirect potable use or sanitary applications.

The tested pilot setting worked without any flaws during the trial with a total duration of 18 months. The major advantage of the VFCW was its capacity to compensate and buffer variations in loadings, which allowed to keep the dimensions of the disinfection system small. Maintenance efforts could be reduced to simple bi-monthly system checking, which is a major advancement compared to other settings described [P1.3, P2.1, P3.1 and P3.2]. As neither the dosing of salt for the chlorine production nor the application of the softener was required to run the station, the treatment and disinfection process could be substantially simplified. It can be operated without any chemical input.

From these results the following suggestions for future system improvement have been derived:

1. Adapt vegetation to reduce ET.
2. Intensify the operation of the VFCW e.g. by forced aeration to further reduce the area required of CWs.
3. Design CWs to treat raw WW instead of polishing only. Through that the combination of CW and OCG may become a complete treatment solution for the reuse of WW in rural regions.

6.4 Publications related to this topic

[P3.1]

Álvarez, J. A., Ávila, C., **Otter, P.**, Kilian, R., Istenič, D., Rolletschek, M., Molle, P., Khalil, N., Ameršek, I., Mishra, V. K., Jorgensen, C., Garfi, A., Carvalho, P., Brix, H. and Arias, C. A. **2017** Constructed wetlands and solar-driven disinfection technologies for sustainable wastewater treatment and reclamation in rural India: SWINGS project. *Water Sci Technol*, wst2017329.

[P3.2]

Mishra, V. K., **Otter, P.**, Shukla, R., Goldmaier, A., Alvarez, J. A., Khalil, N., Avila, C., Arias, C. and Ameršek, I. **2018** Application of horizontal flow constructed wetland and solar driven disinfection technologies for wastewater treatment in India. *Water Practice and Technology*, **13**(3), 469–480.

[P3.3]

Otter, P., Hertel, S., Ansari, J., Lara, E., Cano, R., Arias, C., Gregersen, P., Grischek, T., Benz, F., Goldmaier, A. and Alvarez, J. A. **2020** Disinfection for decentralized wastewater reuse in rural areas through wetlands and solar driven onsite chlorination. *Science of The Total Environment*, **721**, 137595.

7 ECl$_2$ of treatment step for Trace Organic Components

7.1 Concept

Studies conducted world-wide have confirmed the occurrence of pharmaceutical residues in the effluents of WWTPs (Hirsch *et al.* 1999), water streams (Kolpin *et al.* 2002; Schwab *et al.* 2005), surface and groundwaters (GW) (Avisar *et al.* 2009; Ebele *et al.* 2017). Among those TOrCs, pharmaceuticals, personal care products, sweeteners, illicit and non-controlled drugs, complexing agents, nanoparticles, perfluorinated compounds, pesticides, flame retardants, fuel additives and endocrine disrupting compounds have become a matter of concern (Houtman 2010; Montes-Grajales *et al.* 2017). Conventional WW treatment such as activated sludge processes, exhibits limitations in the removal of TOrCs (Mohapatra *et al.* 2016; Tran *et al.* 2018). Therefore, TOrCs are also present in drinking water, especially in developing regions where pharmaceuticals are often produced and discharged in large quantities. In India e.g. ciprofloxacin, sulfamethoxazole, amoxicillin, norfloxacin, and ofloxacin concentrations were found up to 40 times higher in WWTP effluent compared to other countries in Europe, Australia, Asia, and North America (Mohapatra *et al.* 2016; Balakrishna *et al.* 2017). TOrCs are also found in drinking water because conventional drinking water treatment processes such as coagulation/flocculation are not designed to effectively remove TOrCs (Balakrishna *et al.* 2017).

For the removal of TOrCs different advanced oxidation processes (AOPs) have been studied. Most of them are based on ozonation, Fenton oxidation, or UV based AOPs such as UV/H_2O_2. A relative newly studied process is the combination of chlorination with UV radiation (UV/Cl_2) (Sichel *et al.* 2011; Fang *et al.* 2014; Wang *et al.* 2016; Xiang *et al.* 2016; Rott *et al.* 2018). UV/Cl_2 has proven to produce higher amounts of (•OH) radicals compared to UV/H_2O_2 mainly due to higher absorbance of UV light by chlorine compared to H_2O_2 (Watts and Linden 2007; Fang *et al.* 2014). The Cl_2+UV process has proven superior to the UV/H_2O_2 process to degrade a wide variety of compounds such as benzotriazole, carbamazepine and diclofenac (Sichel *et al.* 2011; Rott *et al.* 2018). Sichel *et al.* (2011) also estimated that the specific energy demand of a state-of-the-art UV/H_2O_2 process could be reduced by 30-70 % when applying UV/Cl_2 process.

7.2 Treatment approach

In a pilot study [P5], the dosing of chlorine was substituted by the inline electrolytic production of chlorine and for the first time combined with a UV light source (PURION®

2500 36 W low pressure UV-C reactors). The target of this study was to evaluate whether an UV/ECl_2 combination is in principle capable to reduce TOrCs. Lab tests were successfully conducted to estimate the radical formation from photolyzed chlorine produced by ECl_2 beforehand.

In a field test with Elbe river water an ECl_2 system was equipped with up to two UV lamps to be tested for its capacity to remove selected TOrCs. Further the “electric energy per order” (E_{EO}) of the degradation was calculated. During this test, the specific energy input into the electrolytic cell as well as into the UV radiation was varied by adapting the flow rates, the current at the electrolytic cell and/or the number of lamps (Fig. 20).

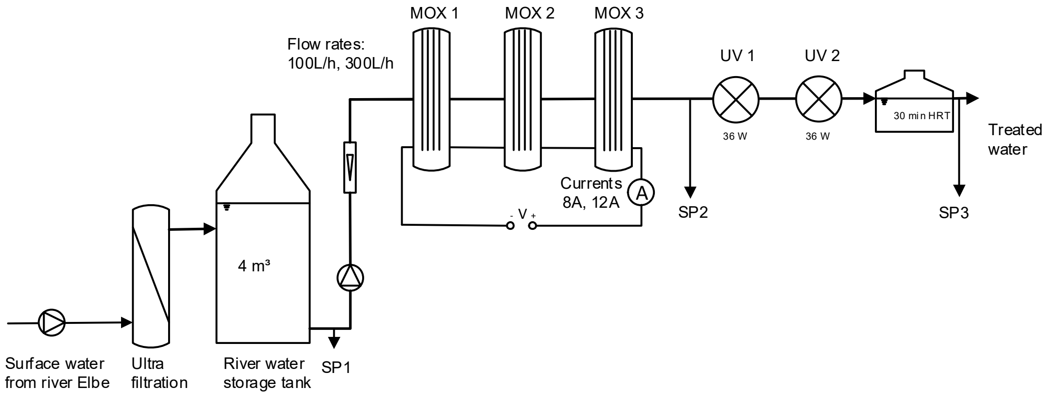


Fig. 20: Pilot system tested with Elbe river water

7.3 Results

The lab tests have shown that the quantity of FAC degraded is proportional to the radicals formed (Fig. 21) and the hydroxyl radical yield accounted for about 64 % per mole of FAC degraded. There was no pH dependency of this yield, which would largely simplify to determine the radical formation by simply monitoring the chlorine degradation during the passage through the UV lamp.

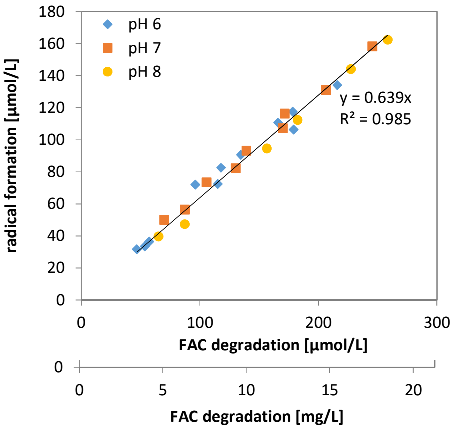
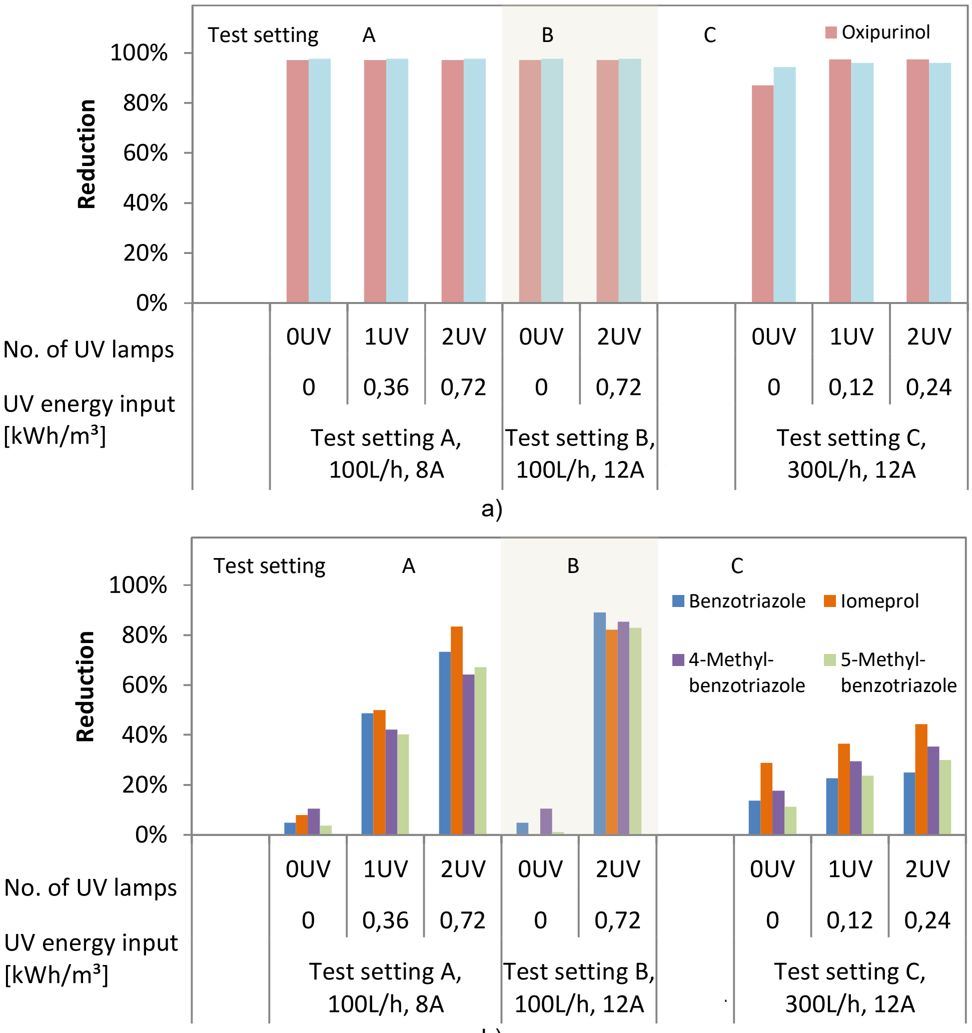


Fig. 21: Radical formation in dependence of FAC demand at different pH (Q = 75 L/h, EC = 400 µS/cm, T = 19 ± 1°C, n = 26)

The field test showed that benzotriazole present in Elbe river water could be degraded by 5 % by ECl_2 alone. In combination with UV the degradation increased to 89 %. Similar results were achieved for 4-methylbenzotriazole, 5-methylbenzotriazole and iomeprol. Oxipurinol and gabapentin were readily degraded by ECl_2 already without the application of UV (Fig. 22).



a)

b)

Fig. 22a and b: Degradation percentages of selected TOrC measured in three different settings during field tests with Elbe river water

Disinfection by-products measured as THMs were maintained below the German drinking water standard of 50 µg/L, provided free residual chlorine concentrations were reduced to the permissible drinking water limits.

The long-term UV/ECl_2 trial was conducted over a period of 10 months during which 1,023 m^3 (3.4 m^3/d) of water were treated. Except for UV lamps cleaning, no maintenance steps were required. The components, and especially the electrolytic cells, did not show any sign of wear by the time the trial was finished. No chemicals for the operation and maintenance of the system were required.

7.4 Conclusion and future prospect

The photolysis of chlorine (UV/ECl_2) was therefore identified as a promising future AOP for the degradation of TOrCs present in drinking water sources. Although promising and done under real field test conditions, the here presented work only represents preliminary lab experiments and a single field trial. Further in-depth analyses are required to confirm those results. This should include the evaluation of the degradation of further TOrCs, the generation of time course data and the analysis of potential forming transformation products originating from the AOP and the evaluation of potentially increased toxicity of the water. From those data design criteria for UV/ECl_2 treatment systems can be derived. Interesting for future applications may also be the treatment of treated WW prior to its discharge into the environment.

7.5 Publication related to this topic

[P5]

Otter, P., Mette, K., Wesch, R., Gerhardt, T., Krüger, F.-M., Goldmaier, A., Benz, F., Malakar, P. and Grischek, T. **2020** Oxidation of Selected Trace Organic Compounds through the Combination of Inline Electro-Chlorination with UV Radiation (UV/ECl2) as Alternative AOP for Decentralized Drinking Water Treatment. *Water*, **12**(11), 3275.

8 Onsite Chlorine Generation as an alternative to ECl_2

8.1 Operational Challenges

During the here conducted field studies, increased levels of hardness in the water have caused substantial operational challenges. At some places even short polarity inversion intervals of ≤ 60 minutes could not completely remove calcareous deposits (Fig. 23). Remaining deposits acted as nucleation centers and the cathodes quickly covered again. The deposits put the overall treatment approach at stake. The operational challenges persisted throughout the presented field tests in WB [P1.3] and Uttarakhand in India [P2.1], as well in adjacent studies in Costa Rica [P1.2]. Only with weekly intensive cleaning and maintenance efforts these trials could be continued. The here conducted studies have shown that for waters containing total hardness levels above 200 mg/L $CaCO_3$ alternative technical solutions must be developed.

Fig. 23a-c: Completely clogged electrolytic cell during pilot trial in WB India [P1.3] and new setting as tested during pilot trial in Almeria Spain [P3.3] with extremely hard waters

Here the application of OCG may pose a feasible alternative to the application of ECl_2. During OCG a chlorine stock solution is produced from a NaCl brine solution. This stock solution is then dosed into the water to be treated.

Due to elevated hardness levels the OCG settings were also applied in some of the here presented case studies: Whereas in the developed setting [P3.3] no addition of NaCl was required, due to sufficiently high chloride concentrations in the raw water (819 ± 132 mg/L), in the treatment systems described in [P4] NaCl is added.

The water necessary to produce the brine hereby makes up only a small portion of the total water volume to be treated. This ratio comprised 4-23 % of the total volume of WW treated [P3.3] and only about 0.1 % for the total volume of drinking water applications. During the WW disinfection case study [P3.3] electrode scaling was prevented through polarity inversion alone, despite the elevated hardness levels in the water. Not even softening of the small portion was required.

8.2 Energetic considerations

To compare the energy requirements to produce chlorine, Tab. 6 summarizes the achieved chlorine production rates and the respective quantities of electricity consumed. The data were derived from lab and field studies [P2.1 and P3.3]. ECl_2 and OCG settings that are operating with different chloride concentrations but equal MOX electrodes are compared.

Tab. 6: Energy requirements of equal MOX electrodes under different operating conditions

Setting		Appli-cation	dosing of NaCl required	Chloride conc. [mg/L]	Flow rate [L/h]	Current [A]	Voltage [V]	Power [W]	Total chlorine [mg/L]	Chlorine production rate [g/h]	Salt yield [%]	Specific energy consumption [kWh/kg]
ECl_2	lab	DW	no	100	75	6	11,5	69,0	17,1	**1,0**	17 %	**50,4**
	Haridwar	DW	no	14±2	220	2-5	12,0	24-60	0,3	**0,7**	2 %	32 - 83
OCG no dosing*	Spain	WW	no	819±132	170	5	4,8	24	151 ± 15	**1,8**	18 %	13,3
OCG	lab	DW	yes	1.300		5	3,9	19	619	**3,1**	48 %	6,3
	lab**	DW	yes	2.600		5	3,1	16	930	**4,3**	36 %	3,8
	lab	DW	yes	5.200		5	2,8	14	958	**4,8**	18 %	**2,9**

* due to high "natural" chloride concentration in WW no NaCl was added ** currently preferred setting for drinking water disinfection

The presented numbers clearly show the difference between ECl_2 and OCG in the energy required for producing chlorine and the chlorine production rates. Whereas in lab studies 50.4 kWh were required to produce one kg of chlorine, only 2.9 kWh/kg were required with OCG. At the same time the chlorine production rate increases from 1.0 to 4.8 g per hour, with the same MOX electrode model. This has not only implications on the energy demand of the setting but also on the economic feasibility and the size of the electricity supply unit for such stations. In case stations are to be powered with solar PV generated electricity, adequate combinations of PV and batteries to operate such a chlorine electrolysis disinfecting system must be determined. Next to the smaller amount of energy required, an additional advantage of OCG is the possibility to shift the chlorine production hours into the daytime, when solar energy is available. This reduces the requirements on energy storage capacities in batteries. Examples of such settings are shown in Tab. 7 for the ECl_2 system operating in Haridwar [P2.1] and a comparable OCG system, where the required amounts of chlorine can be produced within in one daytime hour of operational time.

Tab. 7: Selected PV battery combinations to power OCG and ECl_2 systems

Setting	PV power and battery capacity	Total prod. [kWh/a]	Total cons. [kWh/a]	Input battery [kWh/a]	Output battery [kWh/a]	Excess electricity [kWh/a]; [%]	Unmet load [kWh/a]; [%]	Capacity shortage [kWh/a]; [%]
1. OCG	300 W; 25 Ah	489	195	110	91	275; 56.2	1.77; 0.9	2.01; 1.0
2. OCG	300 W; 50 Ah	489	197	112	93	273; 55.8	0.11; 0.1	0.12; 0.1
3. ECl_2	300 W; 50 Ah	489	422	200	167	33; 6.8	140; 24.9	158; 28.1
4. ECl_2**	900 W; 96 Ah	1,466	561	325	271	850; 58.0	1.09; 0.2	1.23; 0.2

*22 h/day of system operation, power requirements of 24 W to pump 180 L/h and average chlorine dosing of 1 mg/L **PV battery system as installed in Haridwar

The table shows that for the in Haridwar [P2.1] given pilot site the dimensions of the solar power supply setting could be reduced by 70 % for the PV panel size and by 50 % for the battery size when changing from ECl_2 to OCG hereby maintaining or even reducing the periods of unmet load to 0.1 %. These correspond to less than 9 hours per year under the given operation conditions. Fig. 24 and Fig. 25 show the with HOMER (Lilienthal 2009) simulated battery state of charge for the given system settings.

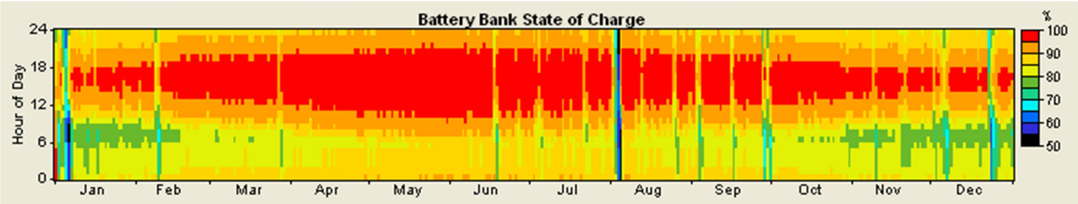


Fig. 24: Battery state of charge for a 300 Wp PV and 50 Ah battery setting to power the OCG system including well pump

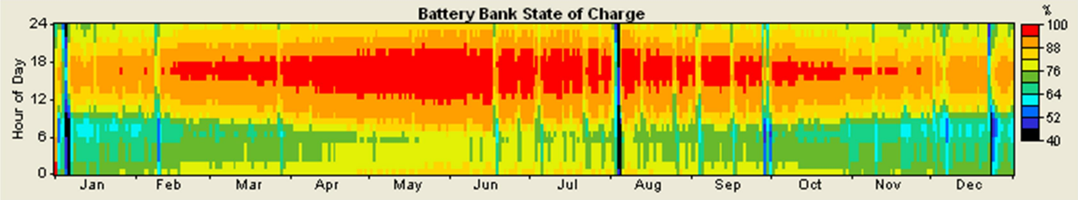


Fig. 25: Battery state of charge for a 900 Wp PV and 96 Ah battery setting to power the ECl_2 system including well pump

Additionally, the following aspects supporting the application of OCG have been identified:

8.2.1 Process control and system design

With OCG the process of chlorine production and dosing is decoupled from water pumping and is not influenced by the raw water quality. This allows to generate constant and known chlorine concentration and simple dosing of chlorine as per raw water requirements. In ECl_2 source water quality has major impact on the chlorine production process, which are hard to evaluate especially in rural regions, with reduced data availability. OCG systems can be designed and installed largely independently from source water conditions.

8.2.2 Formation of inorganic DBPs during storage of stock solution

A potential challenge in OCG operation is the formation of chlorate and perchlorate especially under high current densities (Czarnetzki and Janssen 1992). To evaluate the chlorate production of potential OCG systems lab campaigns were conducted using different market available MOX electrodes. The target of this campaign was to identify operational conditions of an OCG setting at which the electrolytic formation of chlorate is not of concern at the same time allowing for adequate chlorine production rates. For that three different MOX electrodes were used and tested by varying salt concentrations and current densities. Cell "B" is hereby the MOX electrode which was used throughout the trials described here. An excerpt of the generated results is shown in Tab. 8.

Tab. 8: Selected operational parameters and achieved chlorine and chlorate concentrations at a chloride concentration of 2.6 g/L in the brine solution

Cell	Current [A]	Current density [µS/cm²]	Wattage [W]	Chlorine conc. [mg/l]	Chlorine production rate [g/h]	Salt Yield	Energy specific FAC production rate [mg/Wh]	Chlorate conc. [mg/l]	ClO_3/Cl_2	Chlorine added [mg/L]		
										0.5	1.0	2.0
										Resulting ClO_3 concentration [µg/L]		
A	2.0	66.7	9	45	0.9	2%	106	1.2	3%	13	26	52
60 cm²	10.0	333.3	145	156	3.1	6%	22	33.2	21%	106	213	426
	15.0	500.0	288	272	5.4	10%	19	77.3	28%	142	284	568
B	2.0	6	5	61	1.2	2%	235	0.9	1.4%	7	14	29
639 cm²	5.0	16	17	205	4.1	8%	248	1.5	0.7%	4	7	15
	8.0	25	32	328	6.6	13%	205	4.5	1.4%	7	14	27
C	2.0	4	14	75	1.5	3%	109	1.7	2.3%	11	23	46
900 cm²	3.5	8	28	252	5.0	10%	180	3.5	1.4%	7	14	28
	5.0	11	44	368	7.4	14%	167	7.3	2.0%	10	20	40

preferred setting

not permissable in Germany

These results show that cell A is producing excessive amounts of chlorate, which reached up to 28 % of the chlorine concentrations produced in the stock solution. The comparatively high chlorate production was related mainly to the small dimensions of the cell and the resulting high current densities. Depending on the guidelines applied (Tab. 3) the resulting concentrations would lead to critical levels in the disinfected water. Therefore, cell A was not considered for further testing. When using cell B and cell C the chlorate concentrations that would result in the treated water are of no concern. Then, only cell B and cell C were chosen to be used in OCG.

Chlorate and perchlorate also form during the storage of stock solution through the degradation of hypochlorite as described in Eq. 8 and Eq. 9.

$$2\,HOCl + OCl^- \rightarrow ClO_3{}^- + 2H^+ + 2Cl^- \qquad \text{(Eq. 8)}$$

$$OCl^- + ClO_3{}^- \rightarrow ClO_4{}^- + Cl^- \qquad \text{(Eq. 9)}$$

The degradation rate is a function of temperature, concentration, ionic strength, pH and time (Stanford *et al.* 2011; Breytus *et al.* 2017). Models to calculate its concentration in dependence of those factors are available e.g. by (AWWA 2020). Dilution of the chlorine stock solution is the most effective way to reduce chlorate and perchlorate formation during storage. (Stanford *et al.* 2011) e.g., suggest diluting 13 % (124 gCl_2/L) stock solution by a factor of 4 to reduce the perchlorate formation rate by a factor of 36. Storage time should not be longer than 1-2 days if the pH is not corrected to above 11. OCG systems generally produce chlorine solution on demand and have only small storage capacities for about one day. With the in Tab. 8 shown preferred setting, chlorine concentrations of only <1 g/L are anticipated. Therefore, the chlorate formation potential caused by the degradation of chlorine is negligible during OCG.

9 Economic considerations of decentralized drinking water supply schemes

9.1 Background information on drinking water supply in rural developing regions

Due to environmental fragility and poor economic conditions of rural developing regions the supply with safe water in such areas is more challenging compared to urban regions. Most people without access to improved drinking water sources live in rural areas. Water technologies are often designed for urban contexts and do not take the particularities of rural areas in terms of energy efficiency, the use of natural treatment systems and cultural appropriateness into account. Due to this, long-term operation under such challenging conditions of common treatment systems often fails (Dettmar *et al.* 2017; AUC 2019). Public water provision services, as well as already implemented water infrastructures such as water collection points, are often inadequately maintained and in poor shape. In addition to this lack of services, natural water sources such as groundwater and surface water are often contaminated and provide an unreliable supply.

Therefore, site-specific technological approaches tailored to the specific water and energy needs of rural developing areas are required. This is costly, considering the small number of consumers supplied in rural areas.

With water point-based solutions basic drinking water services can be assured if they are reachable within a 30-minute walking round trip from a household. By that they are capable in serving the most remote and vulnerable population and pose an intermittent low cost alternative to household connections, provided the supplied water is adequately treated with regards to existing guidelines (WHO 2019; Hope *et al.* 2020). Water points also allow the future extension of the water supply network to each household and can be an intermediate step to reach the SDG No. 6.

For a successful operation of water point solutions, the participation and collaboration of the community, in which such points are established, is required. This will not only help to reduce the capital investment costs but also ensures consumer buy-in for the water supply and may even allow lower water prices (Jaskolski and Pellegrino 2019). Options for participation are e.g., the construction of water networks, buildings, and reservoirs, using local knowledge and materials. However, community participation during the implementation phase alone is not sufficient and community-run treatment stations easily become defunct as shown e.g. by Das (2013) and Hoque *et al.* (2004). Additionally, financial, and operational management is necessary to operate the water supply scheme on the long-term. Rural public entities and communities are usually not able to finance the required water treatment, supply, and monitoring facilities (Hope *et al.* 2020). At the same time, there is a continuous interest from development partners and social investment funds in supplying financial resources for safe water supply infrastructure, provided the output of such funds can be measured (Koehler 2018; Hope *et al.* 2020). Nevertheless, considering the comparatively small investment required for a community driven rural water supply scheme, a professional economic evaluation as done e.g. by major development banks is usually neither required nor conducted. For investors, NGOs, and communities it is therefore rather difficult to evaluate the economic feasibility and profitability of such small investments. Often financial resources are inefficiently allocated or even wasted. This knowledge gap hampers private investors and NGOs playing a larger role in closing the financing gap for rural water supply schemes.

9.2 Evaluation of economic feasibility of drinking water supply systems

Three drinking water supply projects from Nepal (Devgaun), Egypt (El Heiz) and Tanzania (Rombo) have been evaluated in depth on their economic feasibility to close such gaps [P4]. A prerequisite for site selection was that consumers pay for water via a volume-based tariff. Based on the data examined, it is discussed whether the initially assumed and locally set prices in combination with the water quantities consumed were sufficient to a) cover the O&M costs or b) cover O&M plus investment costs. The presented study [P4] further investigated water prices and the water consumption necessary to permit a profit margin for stations that are implemented as an economic business investment rather than an externally funded development project.

In the study the water treatment system implemented was based on the technological platform offered by AUTARCON GmbH and comprises autonomous units for electrolytic chlorine production either by means of ECl_2 or OCG. The evaluated stations are 100 % powered by solar energy and capable of operating completely independent of an external electricity supply and any added chemicals, except for the addition of sodium chloride (NaCl) in Tanzania and Nepal. In Egypt, the chlorine is produced through ECl_2. In Nepal and Tanzania stock solution is produced through OCG from a NaCl brine solution and then dosed proportionally into the water. Additionally, iron and ammonium were removed in Nepal through aeration and subsequent filtration. In Egypt iron was removed through aeration and subsequent slow sand filtration and additional GSP filtration after ECl_2.

In Nepal cash payments of 20 NPR (7.7 €/m³) were required per 20 L jar. Small volumes of the treated water were given out for free to the students of the school, on which's premises the station was installed. In Egypt and Tanzania water was sold via prepaid water ATM cards at costs of 1-2 Piaster/L (0.7-1.4 €/m³) and 2.5 TSH/L (0.9 €/m³) respectively.

9.3 Economic evaluation

For the evaluation of the economic feasibility, the annual cash flow, and the net present value (NPV) from each project site were calculated, assuming a system lifetime of 15 years and using the initial capital investment costs (I) as well as the present values (PrV) of O&M costs. Projects were considered sustainable when long-term O&M costs can be covered, and profitable when in addition to that the break-even point (BEP) can be reached within five years (payback period). This flow of income is considered as the only revenue for paying O&M and potentially capital investment costs in this study.

Key quantitative and qualitative factors that have an influence on the economic feasibility as well as supplementary recommendations for the successful operation of rural water supply projects in general were derived from interviews conducted with the respective project managers. Those should help project planners to evaluate similar projects more realistically in the future.

9.4 Results

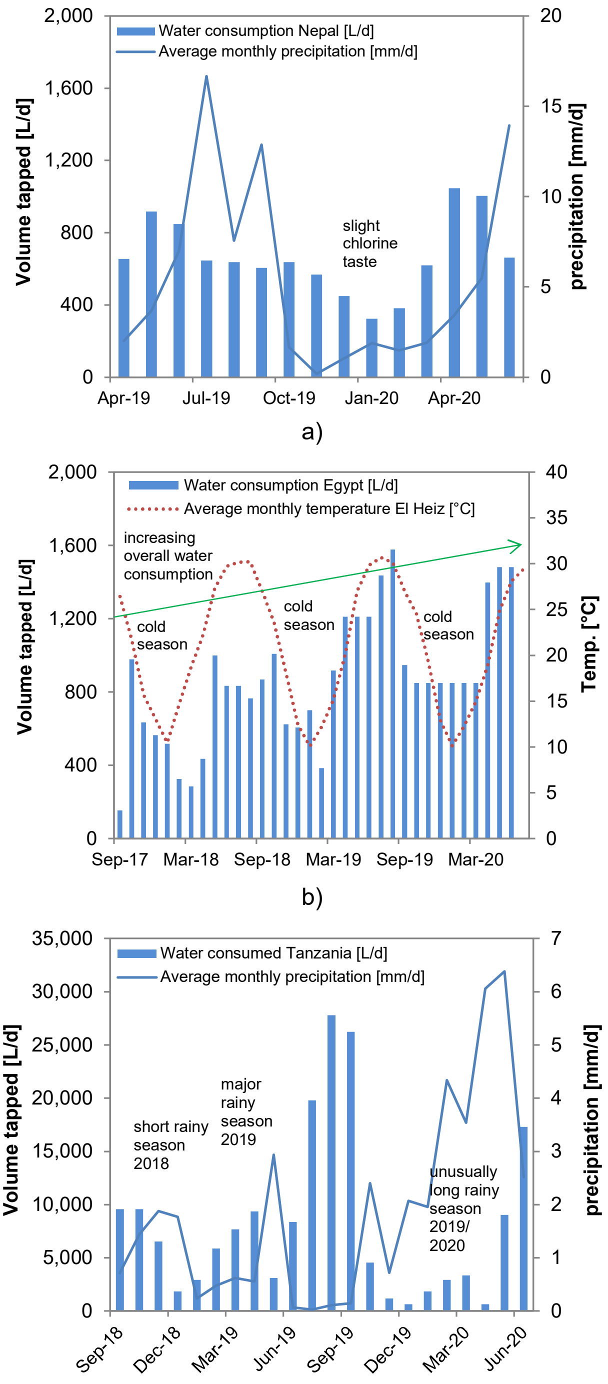
The identified costs and revenues of the stations in Nepal, Egypt, and Tanzania that were required for NPV evaluation are summarized in Tab. 9.

Tab. 9: O&M costs for the presented rural water supply systems

	Nepal	**Egypt**	**Tanzania**
Total investment costs [€]	**24,100**	**23,605**	**44,010**
People served [capita]	2,000	1,500	2,000
Investment costs per capita [€/(cap)]	12.1	15.7	22
Running Costs			
Operation and Maintenance [€/a]	1,235	547	1,004
Spare parts [€/a]	580	394	917
Total running costs [€/a]	1,815	547	1,921
Running costs per capita [€/(a*cap)]	0.91	0.36	0.96

**Reduced customs duty for water treatment systems are assumed for water supply infrastructures

The stations' performance with regard to the water consumption was evaluated over a period of 15, 34 and 22 months in Nepal, Egypt, and Tanzania respectively and the average daily water consumption is shown in Fig. 26.



a)

b)

c)

Fig. 26a-c: Average daily quantity of drinking water tapped in Nepal (a), Egypt (b), and Tanzania (c)

The quantities of water consumed reached with 17 % in Nepal, 30 % in Egypt and 22 % in Tanzania only a portion of the originally by the project planners expected volumes. While in Tanzania the maximum treatment capacity of the station was met at some days during the dry periods, the stations in Nepal and Egypt remained idle most of the time. This low average water consumption had a substantial effect on the revenue generated and the economic feasibility of the projects.

Still, the NPV evaluation (Fig. 27) showed that in Nepal and Tanzania all O&M costs can be covered and even a small surplus was generated. In Egypt, the long-term O&M costs cannot be covered, and the system is running at a loss. Despite the ongoing increasing average water consumption in Egypt over the past years (Fig. 26b) a price increase by a factor of 2.8 is necessary to cover long-term costs. In [P4] this factor was defined as m_{qp} comprising water quantity and price increase.

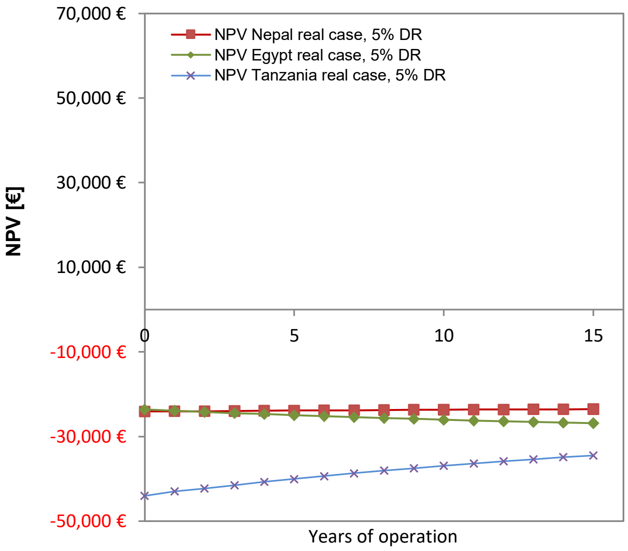


Fig. 27: Net present value of evaluated field cases

The study showed, that the m_{qp} necessary to recover the investments costs are with 6.0, 9.0 and 5.2 for Nepal, Egypt, and Tanzania (Tab. 10), respectively, of rather theoretical nature.

Tab. 10: Required m_{qp} to meet break even within five years or to cover running costs

Break even in 5 years at a 15 % discount rate (investor business case)	**Nepal**	**Egypt**	**Tanzania**
m_{qp}	6	9	5.2
Quantity of water required [m³/d]*	4	7.9	44.8
Revenue through water sales [€/a]**	11,231	8,010	15,426
Total O&M costs [€/a]	3,610	894	1,921
Cash surplus [€/a]**	7,621	7,116	13,506
Break-even [a]	5	5	5

*no price change considered here *** present value at the beginning of the project

Consequently, initially invested capital cannot be recovered and private investors would not be interested in funding such projects. This shows the importance of donor and development bank driven project initiatives.

Additionally, qualitative key factors influencing water volumes consumed, together with supplementary factors supporting long-term system operation have been identified and summarized in Tab. 11.

Tab. 11: Key factors identified to impact the quantity of water sold and supplementary factors identified as beneficial for long-term operation of a rural water supply system

Key factors directly impacting the quantity of water sold
- **Alternative water sources** will limit the consumption of water to drinking water applications only
- **Reliable water supply and water quality** help to create trust in the treatment and supply system and may draw consumers from alternative - less reliable - sources o Removing known and visible impurities such as turbidity, iron precipitates etc. and improving the waters taste, additionally to the required supply of residual disinfectant is more convincing compared to the supply of disinfectant only o **Monitoring** of water treatment system performance and water quality through online devices and manual checking by operators reduces downtimes and increases reliability
- **Awareness** campaigns to inform consumers of a community about the availability of safely disinfected water and the benefits of consuming such
- **Adequate pricing** is crucial but only low water prices may be acceptable in rural settings
Supplementary factors indirectly impacting economic feasibility.
- Sufficient beneficiaries with easy access to a potential water supply system must be available and they must be ready to pay for water - Use of existing abandoned infrastructures, like wells or buildings, should be considered and may reduce investment costs - Participation and cash contributions by consumers can help to reduce investment costs and increase community buy-in of the project. Initial contributions can be e.g., converted to water credits later. Active contribution of community members also helps to spread the information about the availability of safely treated water - Keeping components for likely repairs in stock reduces down-times of systems - Local operators can be encouraged by linking their salary to the quantity of water consumed - Training of local technicians to operate and maintain the treatment system simplifies troubleshooting (in Egypt and Tanzania operators have been taught to install new stations themselves) - Mobile based social media applications simplify exchange of information and allows remote troubleshooting - Below market interest rates are required if projects are funded by loans - Long-term O&M costs of the water supply systems need to be realistically estimated prior to the implementation of a system - The effect of flexible market driven pricing on the revenue generated is promising, but still needs to be evaluated in future projects - Additional sources of income such as CO_2 compensation may support projects otherwise running on a loss

9.5 Conclusion

The study showed that the water volume sold has the highest influence on the economic feasibility of a project. Two out of three evaluated water supply project successfully yielded a cash surplus through the sale of water, which was sufficient to cover O&M cost. However, no site showed a realistic chance of recovering the initial investment costs, within the projected lifetime of the technical infrastructure. According to the reported study drinking water supply in rural regions therefore continues to depend on donors or public funding.

9.6 Publications related to this topic

[P4]

Otter, P., Sattler, W., Grischek, T., Jaskolski, M., Mey, E., Ulmer, N., Grossmann, P., Matthias, F., Malakar, P., Goldmaier, A., Benz, F. and Ndumwa, C. **2020** Economic evaluation of water supply systems operated with solar-driven electro-chlorination in rural regions in Nepal, Egypt and Tanzania. *Water Research*, **187**, 116384.

10 Summary and conclusion

In literature the ECl_2 has been considered as an innovative approach to produce chlorine for decentral disinfection of contaminated water and the supply of residual disinfectant, without the need for the otherwise required chemical dosing. It therefore offers - in theory - an alternative method for the disinfection of water, which can be applied in rural developing regions where direct supply and dosing of disinfectant is challenging. As strong oxidizing agents are commonly used in a variety of water treatment processes, the electrolytic generation of chlorine also offers additional treatment perspectives. While literature on chlorine production mechanisms and the factors influencing the chlorine production at MOX electrodes is available, knowledge on the applicability of ECl_2 under real case conditions and in target regions was missing.

The main task of this dissertation work was to identify, test and evaluate potential water treatment applications for ECl_2 under real case conditions. Scientific assessment of data that have been generated during a multitude of long-term field tests represents the major benefit of this dissertation.

For the disinfection of drinking water and wastewater pilot stations were installed in Haridwar, India and in Almeria, Spain respectively. To test the capacity of ECl_2 in enhancing As(III) oxidation and co-precipitation together with iron hydroxide, pilot stations were installed in Germany, Costa Rica and West Bengal, India. First trials allowed to identify application limits of ECl_2, which required an adaption of the technology during the field campaigns. Therefore ECl_2 was complemented by OCG in some of the here tested pilot systems. To reduce source water quality fluctuations, ECl_2 and OCG systems have been combined and evaluated for the first time with natural treatment steps such as RBF and CWs. Also, for the first time an ECl_2 pilot system installed in Dresden, Germany was combined with an UV lamp to enhance the removal of TOrCs. The target of this trial was to confirm preceding lab results, in which the photolysis of chlorine has resulted in the formation of •OH and •Cl radicals and achieved a substantial degradation of TOrCs. At last, systems that incorporated ECl_2 or OCG as treatment step have been evaluated for their economic feasibility through the assessment of long-term data generated during the operation of such systems in Tanzania, Egypt, and Nepal.

In the presented trials, ECl_2 has proven to be an effective technology to produce oxidizing agent, which can be used for multiple treatment approaches. Especially the combination of ECl_2 with natural treatment systems like RBF has achieved promising results. Here, RBF supplied stable and adequate raw water qualities that allowed an efficient disinfection of the otherwise turbid and fecally polluted Ganga river water. In general, the application of solar PV technology and GSM based data logger have largely simplified the operation and monitoring of the pilot stations. Considering the recent price drops for solar technology and for online devices as well as their simple application, they will also play a major role in future market applications, hereby facilitating system operation and maintenance. During the field trials also local technicians could be trained to maintain and even reinstall such systems whenever required.

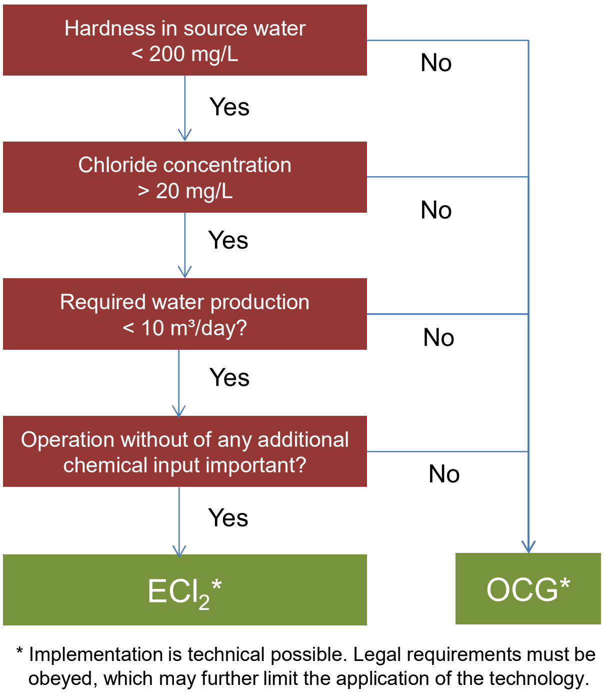
However, the hypotheses stated in chapter 1.2 could be confirmed only in parts as the application of ECl_2 is limited to treatment sites where raw water conditions are favorable. The long-term operation of the treatment, especially in West Bengal and Haridwar, was challenging and required more maintenance than originally hypothesized monthly intervals. Main reasons for that were elevated hardness levels, which quickly caused scaling of the cathodes. Next to extra efforts in maintaining the electrodes, locally higher than permitted current densities reduced electrolytic cell lifetime and increased chlorate formation. Further, scaling prevented the applied control algorithms to adequately adjust the chlorine production.

Another challenge were elevated ammonium levels found in the groundwater at the test site in West Bengal. The ammonium consumed most of the chlorine required to oxidize the arsenic and prevented to evaluate the full potential of the ECl_2 to enhance arsenic removal.

For the pilot tests planned for Spain extremely high hardness and elevated ammonium concentrations required an adaptation of the treatment approach. Instead of ECl_2 production of chlorine, where all the water passes the electrolytic cell, only a small portion of the wastewater to be disinfected was used for chlorine production in a separate chamber. The OCG was then dosed into the water. Due to the high chloride concentrations in the wastewater still sufficient chlorine was produced and the electrolytic cells did not show any scaling throughout the 12-month testing period. In a preceding VFCW, ammonium was completely nitrified offering a low maintenance and low-cost option for polishing treated wastewater for rural regions. The disinfected water did not contain any pathogen indicators and could therefore be used even for indirect potable use, irrigation of public accessible areas and sanitary applications.

The degradation rates of TOrCs during chlorine photolysis are promising, however more testing and data are required to reduce organic DBP formation and reduce specific energy demand. The approach may also be tested for its application in wastewater treatment.

Through the data generated during the field tests the range of applications for ECl_2 had to be reduced. The production of chlorine stock solution (OCG) has been proven to be generally better suited for the here tested applications. The pilot trials have shown that the only advantage left for the field application of ECl_2 is that it does not require any supply of chemicals. Once the dosing of NaCl is required, the use of OCG is preferred as it leads to higher salt yields and reduced energy requirements as described in chapter 8.2. Whenever the application of ECl_2 is required, the selection pathway presented in Fig. 28 will assure favorable operation conditions.



* Implementation is technical possible. Legal requirements must be obeyed, which may further limit the application of the technology.

Fig. 28: Selection pathway for ECl_2 and OCG for the evaluated treatment systems

The detailed economic evaluation of existing ECl_2 and OCG treatment stations has shown that through selling the treated water the stations are able to cover their long-term O&M costs even in very low-income settings. Prerequisite for this was a largely autonomous and nearly maintenance free operation of the applied system. Beyond this, the most critical parameters that have an influence on the economic feasibility were the water quantity consumed and the set water price. To recover the investment costs of the evaluated water treatment and supply systems, it is required to operate the stations close to their design capacity. At the selected study sites the systems were operated at 17-30% of their design capacity only.

The findings of this dissertation work have led to the development of several treatment modules already implemented to safely treat contaminated water in rural developing regions. The technology is also further tested for its potential to allow efficient WW reuse. The dissertation presents a comprehensive, science-based technical and economic evaluation of various ECl_2 and OCG systems and its applications in water treatment in a capacity range of 2-200 m^3/day.

11 Outlook

The conducted lab and field studies have revealed the following future research aspects:

Ammonium has been identified as a common pollutant in drinking water sources as well as in treated WW. To permit efficient disinfection with FAC and oxidation of e.g. As(III) its removal is a prerequisite and has only been partly studied here. More comprehensive studies that would lead to an optional module for its integration prior the disinfection are required. For the arsenic removal approach presented here, this would mean that most of the iron required for co-precipitation may already be precipitated and filtered before the arsenic is fully oxidized and an additional adsorption unit for arsenic may become necessary.

Also the handling of arsenic containing sludge remains to be of concern in arsenic removal. In literature given approaches dealing e.g., with the fixation into concrete used for construction have not been largely implemented and more sustainable solutions are required. At the end safe disposal of the sludge may be the only feasible option.

The measurement of ORP has proven to be useful in determining if sufficient oxidant is supplied to the treatment system. A direct conversion into a chlorine concentration is difficult and possible only after onsite calibration, where the water quality does not change. With changing raw water conditions, the ORP reading becomes inaccurate. Here the application of amperometric free and total chlorine probes offers a more precise reading and their market prices have been drastically reduced in the past. However, the applicability of such probes and their additional maintenance requirements must be evaluated before application in rural developing regions.

The lab and field test studies with regards to photolysis of chlorine through UV/ECl_2 has shown the potential of this technology to oxidize a wide variety of TOrCs. However, more intensive sampling and testing is required to confirm and expand the results of these studies. Additionally, the application of UV/ECl_2 or UV/OCG may be an interesting option for the oxidation of TOrCs in treated WW. In any case the formation of transformation products and a potential increase of toxicity of the water must be evaluated.

During this dissertation work the economic feasibility of drinking water systems and potential business models have been evaluated. Future research should also concentrate on the development of business models, which enhance the reuse of the WW.

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13 Publications with first authorship

The numbering of the publications follows the sequence of their publication.

13.1 [P1.3] Arsenic Removal

Otter, P., Malakar, P., Jana, B., Grischek, T., Benz, F., Goldmaier, A., Feistel, U., Jana, J., Lahiri, S. and Alvarez, J. **2017** Arsenic Removal from Groundwater by Solar Driven Inline-Electrolytic Induced Co-Precipitation and Filtration—A Long Term Field Test Conducted in West Bengal. *International Journal of Environmental Research and Public Health (IJERPH)*, **14**(10), 1167.

13.2 [P2.1] Combination of RBF and Solar-driven electro-Chlorination

Otter, P., Malakar, P., Sandhu, C., Grischek, T., Sharma, S., Kimothi, P., Nüske, G., Wagner, M., Goldmaier, A. and Benz, F. **2019** Combination of River Bank Filtration and Solar-driven Electro-Chlorination Assuring Safe Drinking Water Supply for River Bound Communities in India. *Water*, **11**(1), 122.

13.3 [P3.3] Disinfection for decentralized wastewater reuse in rural areas

Otter, P., Hertel, S., Ansari, J., Lara, E., Cano, R., Arias, C., Gregersen, P., Grischek, T., Benz, F., Goldmaier, A. and Alvarez, J. A. **2020** Disinfection for decentralized wastewater reuse in rural areas through wetlands and solar driven onsite chlorination. *Science of The Total Environment*, **721**, 137595.

13.4 [P4] Economic evaluation of water supply systems

Otter, P., Sattler, W., Grischek, T., Jaskolski, M., Mey, E., Ulmer, N., Grossmann, P., Matthias, F., Malakar, P., Goldmaier, A., Benz, F. and Ndumwa, C. **2020** Economic evaluation of water supply systems operated with solar-driven electro-chlorination in rural regions in Nepal, Egypt and Tanzania. *Water Research*, **187**, 116384.

13.5 [P5] Oxidation of Selected Trace Organic Compounds

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14 Publications with co-authorship

[P1.1]

Feistel, U., **Otter, P**., Kunz, S., Grischek, T. and Feller, J. **2016** Field tests of a small pilot plant for the removal of arsenic in groundwater using coagulation and filtering. *Journal of Water Process Engineering*, **14**, 77–85.

[P1.2]

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[P3.1]

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[P3.2]

Mishra, V. K., **Otter, P.**, Shukla, R., Goldmaier, A., Alvarez, J. A., Khalil, N., Avila, C., Arias, C. and Ameršek, I. **2018** Application of horizontal flow constructed wetland and solar driven disinfection technologies for wastewater treatment in India. *Water Practice and Technology*, **13**(3), 469–480.