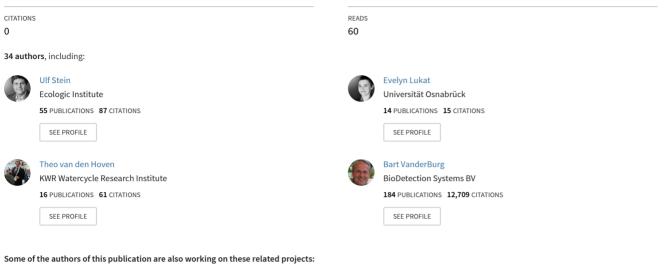
See discussions, stats, and author profiles for this publication at: https://www.researchgate.net/publication/329809963

Challenges and technological approaches for tackling emerging contaminants in drinking and wastewater

Article · March 2018







TECHNEAU View project

Saph Pani View project



Demonstration of promising technologies to address emerging pollutants in water and waste water

Challenges and technological approaches for tackling emerging contaminants in drinking and wastewater

Authors:

Ulf Stein¹, Evelyn Lukat², Theo van den Hoven³, Gerard van den Berg³, Anna Szendrenyi¹, Erwin Beerendonk³, Bart van der Burg⁴, Marc Bourgin⁵, Roberta Caris-Hofman³, Thomas Gross⁶, Stephan Hannappel⁷, Armelle Hebert⁸, Marta Hernández⁹, Hein de Jonge¹⁰, Cornelia Kienle¹¹, Jörg Gebhardt¹², Anna Kounina¹³, Beatriz de la Loma Gonzalez³, Christa S. McArdell⁵, Daniel Mutz¹⁴, Ron van der Oost¹⁵, Miranda Pieron³, Christian Remy¹⁴, Eszter Simon¹¹, Christoph Sprenger¹⁴, Ester Vilanova¹⁶, Kristina Wencki¹⁷, Mariëlle van der Zouwen³, Christoph Hugi⁶, Christopher Oberschelp¹⁸, Merijn Schriks¹⁹, Kirsten Baken³, Hannes Schritt¹

Keywords: Emerging contaminants, innovation, treatment technologies, drinking water, wastewater

¹ Ecologic Institute, Pfalzburger Str. 43/44, 10717 Berlin, Germany

² Institute of Environmental Systems Research, Osnabrueck University, Barbarastr. 12, 49069 Osnabrueck, Germany

³ KWR Watercycle Research Institute, Groningenhaven 7, 3433 PE Nieuwegein, The Netherlands

⁴ BioDetection Systems, Science Park 406, 1098 XH Amsterdam, The Netherlands

⁵ Eawag, Swiss Federal Institute of Aquatic Science and Technology, CH-8600 Duebendorf, Switzerland

⁶ Institute for Ecopreneurship, School of Life Sciences, University of Applied Sciences and Arts Northwestern Switzerland (FHNW), Gründenstr. 40, CH-4132 Muttenz, Switzerland

⁷ Hydor Consult GmbH, Am Borsigturm 40, 13507 Berlin, Germany

⁸ Veolia Environment Research and Innovation, 10 rue Jacques Daguerre, 92500 Rueil Malmaison, France

⁹ CETaqua Andalucía, Calle Severo Ochoa 7, 29590 Málaga, Spain

¹⁰ Dunea Duin & Water, Plein van de Verenigde Naties 11-15, 2719 EG Zoetermeer, The Netherlands

¹¹ Swiss Centre for Applied Ecotoxicology Eawag-EPFL, Überlandstrasse 133, 8600 Dübendorf, Switzerland

¹² Aquatune, In den Wingerten 13, 65623 Hahnstätten, Germany

¹³ Quantis, EPFL Innovation Park, Bât. D, 1015 Lausanne, Switzerland

¹⁴ Kompetenzzentrum Wasser Berlin, Cicerostr. 24, 10709 Berlin, Germany

¹⁵ Waternet, Korte Ouderkerkerdijk 7, 1096 AC Amsterdam, The Netherlands

¹⁶ Amphos 21 Consulting SL, Passeig de Garcia Fària, 49, 08019 Barcelona, Spain

¹⁷ IWW Zentrum Wasser, Moritzstr. 26, 45476 Mülheim an der Ruhr, Germany

¹⁸ ETH Zürich, Rämistr. 101, 8092 Zürich, Switzerland

¹⁹ Vitens N.V., Oude Veerweg 1, 8001 BE Zwolle, The Netherlands

Abstract:

In recent decades, emerging contaminants (ECs) have surfaced as one of the key environmental problems threatening ecosystems and public health. Most emerging contaminants are present in low concentrations, and therefore often remain undetected and are also referred to as 'micropollutants'. Despite this, many ECs raise considerable concerns regarding their impacts on human and environmental health. DEMEAU (Demonstration of promising technologies to address emerging contaminants in water and wastewater), a European Seventh Framework Programme (EU-FP7, 2013-2015) project, aimed to tackle ECs in drinking and wastewater by advancing the uptake of knowledge, prototypes, practices and removal technologies. The project followed a solutions-oriented approach using applied research and demonstration sites, and explored four promising technologies for EC removal and/or degradation: Managed Aquifer Recharge (MAR), Hybrid Ceramic Membrane Filtration (HCMF), Automatic Neural Net Control Systems (ANCS) and Advanced Oxidation Techniques (AOT). Furthermore, Bioassays (BA) were investigated as an effect-based monitoring tool. This article shares new findings for each approach and their potential for widespread integration in the drinking- and wastewater sector. Research results from DEMEAU demonstration sites show that opportunities for synergies among these developments offer the most promising and effective methods for tackling ECs in the water sector.

1 Introduction and objective

Point and diffuse chemical pollution stemming from agriculture (e.g. pesticides), human population (e.g. pharmaceuticals), domestic usage, and industry (e.g. perfluorinated compounds) has posed new challenges to the drinking and wastewater sector over the last decades. Coupled with demographic changes, climate change and an aging and deteriorating water infrastructure, research and innovation in the water sector has become critical to ensure the long-term sustainability and quality of water resources.

Emerging contaminants are substances that are not yet included in routine monitoring e.g. within the EU Water Framework Directive. They have surfaced as contaminants of concern due to their unknown long-term effects on ecosystem and human health [1]. There is already substantial knowledge on the negative impacts of endocrine disruptors on (aquatic) ecosystems, stemming e.g. from pharmaceuticals, industrial chemicals and pesticides from agricultural and urban runoff [2,3]. For example, it is also known that ECs encompass a broader range of chemical agents that enter our environment (e.g. biocides leaching from house paint). Currently, there is little conclusive information on the whole suite of ECs and their impacts on human and ecosystem health. However, Chèvre and Erkman [4] estimated that 100,000 substances are allowed on the European market that can potentially be released into the environment, in addition to non-authorized compounds (persistent substances that used to be allowed or substances transported from other world continents).

In Europe, apart from the 45 priority substances that are outlined in its key piece of water legislation, the Water Framework Directive (WFD); EU Directive [5], the majority of ECs remain undiscovered as many known and especially new or unknown substances are not part of current monitoring routines despite that there are analytical methods available for discovering such substances (e.g. [6,7]). This gap in both monitoring and knowledge regarding health effects makes precautionary action essential, which is embodied in the risk management approach of the WFD.

To manage the potential risk posed by ECs to human and ecosystem health, they require further scrutiny. Several technologies that remove or eliminate ECs in the drinking and wastewater sector have been developed but are still lacking broader implementation across the EU. Switzerland is an exception, where an amendment of the Water Protection Act in January 2016 now requires the implementation of an additional treatment step to remove ECs at certain wastewater treatment plants [8,9].

A better understanding of the link between early biological responses to chemical exposure of populations and communities is critical to closing this gap along with improved knowledge transfer and better science communication among key stakeholders [10]. The DEMEAU project has begun to address these gaps in knowledge and practice by developing and implementing the earlier mentioned four technologies and *in vitro* bioassays in demonstration sites across Europe. The project approach was iterative and interactive, using applied research to involve key stakeholders, including scientists, the private sector, and water utilities at the science-policy interface in order to facilitate uptake of the promising technologies, where appropriate. To this end, DEMEAU advanced new approaches to monitoring, achieving and maintaining a good chemical status of European water bodies. It also explored effect-based approaches to be applied in monitoring practices to highlight opportunities for improved coherence between water quality research (specifically targeting ECs), water regulation (specifically the Groundwater and Drinking Water Directive), and the drinking water and wastewater sector.

2 Methods

2.1 Technologies studied and case studies

ECs occurrence in drinking water is dependent on a variety of factors including the potential EC sources in a catchment, the water source, the water quality highly influenced by weather patterns, production cycles, and/or the abstraction of water for other uses. Consequently, tests must be performed to determine whether further water treatment in addition to standard routines is required.

Within DEMEAU, several promising approaches related to EC removal, process optimization and monitoring have been applied and tested. Technologies that have the ability to remove ECs include: Managed Aquifer Recharge (MAR), Hybrid Ceramic Membrane Filtration (HCMF), and oxidative techniques, including Advanced Oxidation Techniques (AOT). They were chosen due to their specific characteristics: MAR for its potential as natural low-energy system for EC removal but with difficulties to quantify and control EC removal efficiency; HCMF as a compact system combining adsorption and robust new membrane filtration; and AOT as an advanced system with proven technology (ozone, UV) for full-scale application. Additionally, Automated Neural Net Control Systems (ANCS) were applied to optimize technologies such as membrane filtration in order to make these technologies more environmentally and economically efficient. Cell-based (*in vitro*) bioassays were utilized for their potential to be included in routine monitoring practices that assess the presence of ECs and thus to gain an impression of the removal efficiency of the treatment techniques (avoiding the need to measure specific target substances).

A number of case studies (Table 1) represent various stages along the innovation life cycle from the lab to full-scale application. They were conducted under different regulatory framework conditions in several countries, including Germany, Spain, the Netherlands and Switzerland.

Case study	Current system	DEMEAU study	Location	Scale
<u>MAR1:</u> Organic reactive layer	<u>Groundwater recharge:</u>	Addition of an organic layer for improved biological EC degradation	Sant Vicenç dels Horts close to Barcelona (Spain)	Pilot
MAR2: AOP pre- treatment	Drinking water: Dune infiltration of pre-treated (coagulation, flocculation and sedimentation) surface water for later (min. residence time of 20 days) abstraction	Pre-treatment of influent water by ozonation followed by hydrogen peroxide (O ₃ /H ₂ O ₂) and in addition with UV (O ₃ /H ₂ O ₂ /UV)	DWTP of Dunea, the Netherlands	Pilot
HCMF1: Advanced wastewater treatment	<u>Wastewater:</u> Secondary municipal wastewater treatment	Ceramic membrane reactor with powdered activated carbon (PAC)	WWTP near Basel, Switzerland	Pilot
<u>OT1:</u> Advanced wastewater treatment	<u>Wastewater:</u> Secondary municipal wastewater treatment	Ozonation	WWTP Neugut, Switzerland	Full

Table 1 A selection of case studies conducted in DEMEAU, adapted from Gross et al. [11].

BA1: Effect- Drinking wa based water treatm monitoring

Drinking water: Drinking water treatment plant

Screening of possible effects using *in vitro* bioassays Waternet DWTP Pilot near Amsterdam, the Netherlands

2.2 Environmental, economic and societal unique selling propositions and recommendations for impact

To study environmental and economic benefits and potential trade-offs of the technologies investigated in DEMEAU, Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) was performed in the case studies. These results were then used to formulate environmental and economic unique selling propositions (USPs) for each technology. In addition, societal drivers and barriers for technology uptake have been investigated with in-depth involvement of relevant stakeholders in workshops and surveys along the different technology development stages. Finally, USPs were used to derive recommendations on how to improve uptake of these technologies in the water sector and enhance their impact to address the problem of ECs. The results of LCA, LCC and drivers and barriers analysis are only briefly described here, whereas more details are available in the related DEMEAU deliverables available online. [12]

LCA (Figure 1) was used to quantify potential environmental impacts and benefits of each of the technologies assessed in DEMEAU in comparison to existing reference technologies. The comparison was based on process data for EC removal efficiency measured on site or from literature, and energy and chemicals inputs as well as infrastructure collected for each site. Using LCA databases for background information such as the production of electricity, chemicals and materials, as well as the resource extraction and emissions into the environment occurring during these production processes, the overall environmental impact has been calculated as a set of environmental indicators with LCA software. While the effort for this additional water treatment step is mainly reflected on primary energy demand and related emissions (e.g. greenhouse gases), potential benefits of EC removal were characterized by assessing the avoided human toxicity and ecotoxicity impact of the released water.

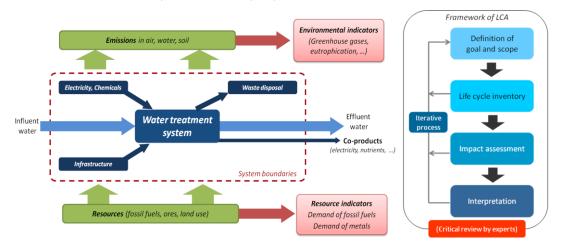


Figure 1: Concept for LCA assessment of water technologies. Left panel: typical system boundaries of an LCA for a water treatment system; right panel: framework of LCA according to ISO 14040/44

One of the challenges DEMEAU encountered with regard to LCA application was (1) the limited number of possibly prevalent ECs actually monitored and measured at the different case study sites, and (2) the lack of specific LCA characterization factors for assessing their potential environmental impact in terms of human toxicity and ecotoxicity. For the first limitation, an extrapolation to the full EC load has been performed to prevent an underestimation of the total (eco)toxicological improvement potential in the water sample, as more ECs have been detected than those monitored. In order to tackle the second limitation, new characterization factors for nine selected organic ECs monitored within the project were calculated based on their toxicological and physico-chemical data, in alignment with the USEtox® consensus

model for toxicity assessment in LCA [13]. These new factors thus enabled an estimate of potentially avoided toxic impacts due to the elimination of monitored ECs with DEMEAU technologies, completed by an extrapolation of the full load toxicity removal that has a high uncertainty.

LCC considers the real costs of applying the technology from a water treatment operator's perspective (i.e. prices for analysis to pay to an external lab) instead of assessing the full life cycle costs of bioassays due to limitations in cost data for lab equipment, chemicals and personnel.

The social drivers and barriers analysis conducted for the same case studies as LCA and LCC provided insights into enabling or constraining factors for market uptake. DEMEAU identified capacity gaps beyond the actual implementation of the technologies encompassing four different levels: (i) contextual, such as policies and regulations; (ii) inter-organizational, such as relationships, agreements and consultative networks among stakeholders; (iii) intra-organizational, such as organizational culture, procedures and resources within organizations; and (iv) individual such as relevant knowledge, skills and motivation of involved individuals. Initially, a drivers and barriers analysis was conducted using online surveys among stakeholders from the selected case studies. Results helped to identify at which level within the innovation process different stakeholders were most active, and revealed perceived drivers and barriers for the wider application of the technologies. Subsequent in-depth drivers and barriers assessment workshops and interviews helped to confront different stakeholder groups with these drivers, barriers and capacity gaps and to define possible ways to overcome barriers.

In the following sections, the five technologies are explored in depth, including an assessment of their potential capacity to tackle ECs, applications of their use via case studies and an assessment of their environmental and economic effectiveness via life cycle analysis and costs.

3 Results and discussion: Studied technologies to deal with emerging contaminants

3.1 Managed Aquifer Recharge (MAR)

MAR describes the intentional recharge (and storage) of water into an aquifer for subsequent recovery and/or for environmental benefits. MAR is applicable for drinking and wastewater treatment, and is often used in combination with additional treatment systems.

MAR has been investigated in a variety of R&D projects and many installations in Europe have been in operation for decades [14]. There is, however, a certain reluctance to install new sites despite that the EU WFD mentions MAR as a possible supplementary measure to achieve good quantitative and qualitative groundwater status. This is partly due to the fact that certain organic trace substances persist in the subsurface and because the efficiency of the subsurface treatment largely depends on site specific characteristics (e.g. sorption capacity, availability of organic matter).

3.1.1 MAR's readiness to tackle emerging contaminants

Removal of ECs during MAR occurs primarily as a result of sorption to organic matter and microbial transformation. Based on a literature review by Vilanova et al. [15], the removal of selected emerging compounds is classified in relation to the predominant redox condition and the residence time in the subsurface (Figure 2). Other factors such as temperature also influence the degradation rates and chemical structure of the ECs, but redox and residence time are found to be most important.

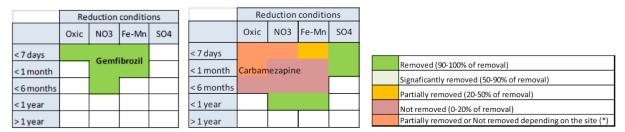


Figure 2: Example of removal matrix of emerging contaminants during subsurface passage (modified from Vilanova et al. [15]).

Due to the redox dependant degradation of some contaminants, only MAR systems comprised of an oxic to anoxic redox sequence ensure maximum attenuation efficiency. Long soil-aquifer passage and high residence time favors interaction with sediments and the communities of microorganisms in the porous media, allowing for removal of contaminants through natural processes.

3.1.2 Success stories of MAR systems

To promote the application of MAR systems and to support operators and authorities to design and operate new MAR systems in compliance with European Water Directives while minimizing energy consumption and costs, DEMEAU developed and validated various supportive tools in the 'DEMEAU MAR²⁰ Toolbox:

- Protocol for soil-column experiments assessing fate and transport of trace organics
- Soil-column study protocol to assess the fate of emerging contaminants under MAR conditions
- Hydraulic characterization of managed aquifer recharge sites by tracer techniques

²⁰ The toolbox includes illustrative case studies (http://demeau-fp7.eu/toolbox).

- Approaches to assess the long-term impact on ambient groundwater
- Risk Assessment methodologies
- Design criteria, including pre-treatment options

With the knowledge of source water and intended end-use, the most appropriate pre-treatment methods and their removal efficiencies for contaminants of main concern as well as removal efficiencies in the MAR systems can now be assessed by using the DEMEAU MAR toolbox.

3.1.3 Environmental, economic and societal considerations of MAR case studies

MAR can substitute or complement water storage, treatment, transfer and supply schemes [16]. Its capacity to remove ECs depends on a number of factors including e.g. MAR type, aquifer type, redox conditions, organic carbon content in the water, residence time, EC concentrations and temperature [15]. Two case studies in DEMEAU aimed to increase their natural capacity to remove ECs: in the case study MAR1 (Table 1) through the addition of an organic reactive layer in pond infiltration, and in MAR2 (Table 1) through AOP pre-treatment. Further AOP case studies are described in chapter 4.3.

The addition of an organic layer in MAR pond infiltration can improve the quality of infiltrated water by removing residual phosphorus and ECs, thus decreasing freshwater eutrophication and human and ecotoxicity. In case study MAR1, Carbamazepine and Sulfamethoxazole are contained in influent river water at low concentrations and are expected to be removed by ca. -10% and -93% compared to influent by an organic reactive layer, based on results from laboratory soil column tests [17]. Due to very small concentrations of target ECs in the influent water, human and ecotoxicological impacts are similar with or without reactive layer in this specific case study. The energy demand and related carbon footprint of MAR pond infiltration would be increased by 40% per unit infiltrated water with a reactive organic layer, but is still very low compared to other more technical water treatment processes [12]. Reflecting the low resource demand during operation, the increase in life cycle costs would be negligible.

In case study MAR2, a pilot-scale pre-treatment before MAR dune infiltration using AOP was tested. Two different AOP setups consisted of ozonation followed by hydrogen peroxide (O_3/H_2O_2) and, in addition, with UV $(O_3/H_2O_2/UV)$. The pre-treatment reduced ECs susceptible to ozone and/or UV and subsequent biological processes in the dunes can further degrade oxidation products. The pilot-scale AOP pre-treatment reduced sulfamethoxazole by -80% and iopromide by -27% compared to the current system, in which bezafibrate, carbamazepine, diclofenac and metoprolol were already reduced below detection limits. Energy for ozonation and UV are the main considerations for environmental and cost impacts. In this example, the AOP pre-treatment would increase the carbon footprint by around +23% without UV and by +63% with UV per unit treated water, compared to the current system. If implemented at full scale, life cycle costs per unit treated water are expected to be +15% higher with O_3/H_2O_2 and +35% with $O_3/H_2O_2/UV$, compared to the current system. Water transport via pumping was the main energy and cost factor in the current system (64% of the energy demand), thus if only water treatment was considered fractions would be proportionally higher.

Online stakeholder interviews [10] showed that public opinion is considered a major barrier by the stakeholders in both MAR case studies, due to interventions changing the physical environment (e.g., pond construction) and concerns about environmental degradation. Particularly the potential (but not implemented) use of reclaimed water (i.e., pre-treated wastewater) has raised opposition in case studies and authorities from the start has been important and effective in both MAR case studies and authorities could be convinced about net beneficial effects on the environment. Probably one of the most significant barriers is the lack of regulatory embeddedness of MAR. In addition, MAR implementation is very context specific depending on type of source water, space, aquifer properties etc., making the establishment of general policies and regulations difficult.

3.1.4 MAR's USPs and recommendations for impact

Based on the preceding considerations, the following environmental and economic USPs can be derived (adapted from Gross et al. [11]):

- MAR fulfils multiple objectives which cannot be replaced by a single technological system, including replenishment of groundwater resources, water storage, water quality improvements and water distribution.
- Dune and pond infiltration are multi-functional barriers for contaminants with low energy and chemical needs.
- AOP pre-treatment can mitigate potential risks from ECs, taking advantage of biological processes for degradation of oxidation products.
- An additional reactive organic layer in pond infiltration can provide water quality improvements, eliminating some ECs with low additional energy needs.

The following main recommendations for impact can be derived (adapted from Gross et al. [18]):

- Administration and policy level: Collaborate on realistic guidelines across involved sectors as well as coordination and dialogue between administrative levels (local, regional to national) and the public from the start of MAR planning.
- Scientific community and technology developers: Take an active role in knowledge transfer between sites, and where necessary utilize and adapt emerging tools, such as e.g. the 'decision tree for MAR impact evaluation' [15] developed in DEMEAU. Disseminate of research results targeting non-scientists and non-experts to help involving local administration, NGOs and the public.
- Utilities: Collaborate with (applied) research and share experiences.

3.2 Hybrid Ceramic Membrane Filtration in water treatment

Hybrid Ceramic Membrane Filtration (HCMF) is a combination of different water treatment technologies with Ceramic Membrane Filtration (CMF). Different combinations were assessed within the DEMEAU project.

Ceramic membranes offer several advantages over polymeric membranes such as better mechanical and chemical stability, better performance and a longer lifetime resulting in lower energy consumption, less waste and a more sustainable operation. In previous EU-projects, the benefits of (hybrid) ceramic membrane filtration were proven on laboratory and pilot-scale for various applications [19].

Until recently, the uptake of ceramic membranes was hampered by relatively high investment costs [20]. These costs could be reduced dramatically because of recent innovations such as hybrid ceramic membrane systems, innovations in reactor concepts [21], developments in automatic process control and integrity monitoring.

3.2.1 Readiness to tackle emerging contaminants

In hybrid membrane processes, powdered activated carbon (PAC) adsorption can be combined with membrane filtration to remove emerging contaminants from drinking water or wastewater. Such hybrid filtration systems have been shown to provide a very effective broadband elimination of these contaminants. In a pilot plant combining PAC adsorption with subsequent ultrafiltration (PAC/UF) or sand filtration at a wastewater treatment plant in Switzerland, more than 80% of over 70 potentially problematic emerging contaminants could be removed at a PAC dose between 10 and 20 mg L⁻¹ [22]. The removal of emerging contaminants is primarily due to the adsorption of these contaminants to PAC and subsequent separation of PAC from the water [22]. Sedimentation and sand filtration are more common PAC separation steps; however membrane filtration has the advantages of complete PAC and bacteria retention, high virus removal and low space requirements [23].

Ceramic membrane systems can achieve a stable operation and performance under high filtration flux rates, high feed water recovery rates and less chemical cleaning needs when compared to polymeric membranes. The strength of the ceramic membranes allows high backwash pressure to provide very good backwash efficiency and makes them furthermore resistant to chemical pre-treatment of the water with oxidants [24]. This would also enhance opportunities to combine oxidative treatment with hybrid membrane filtration. In addition, the appropriate quality of filtrate can be stably obtained without the risk of membrane breakage during a long lifetime (estimated over 10 years according to manufacturer information).

3.2.2 Success stories

DEMEAU contributed to the design and validation of the top and bottom plates of the CeraMac[®] reactor, a new ceramic membrane filtration system. Extensive testing showed that the reactor is stable and ready for implementation in the full-scale drinking water production plant in Andijk (the Netherlands). Currently it is being tested before becoming operable.

To remove ECs from water, ceramic ultrafiltration membranes have to be combined with other treatment steps such as adsorption on activated carbon, creating Hybrid Ceramic Membrane Systems (HCMF). Pilot testing of HCMF at WWTPs in the region of Basel (Switzerland) and Almelo (The Netherlands) showed that EC removal from municipal wastewater was between 50 to 95% for most of the target ECs. Removal efficiency was particularly high for non-polar compounds (benzotriazole, carbamazepine and diclofenac) with 80 to 95 % [23]. Powdered activated carbon (PAC) was a crucial addition to improve removal of ECs and membrane fouling. Bioassays showed that HCMF was capable to remove up to 90% of the compounds triggering response in bioassays. [25]

3.2.3 Environmental, economic and societal considerations of a HCMF case study

HCMF combines an adsorbent such as PAC with ceramic membrane filtration to remove a broad spectrum of ECs from drinking water or pre-treated wastewater at a removal efficiency of more than 80% of ECs found in Swiss wastewater at a PAC dose of ca. 15 mg L⁻¹. Compared with more common polymeric membranes, ceramic ones have generally a higher mechanical strength and durability, higher chemical resistance, higher permeability and longer lifespans. On the other hand, higher investment costs have limited their application in water treatment [20].

Advanced wastewater treatment with HCMF reduces a broad range of ECs. In the HCMF1 pilot-case study (Table 1), hybrid ceramic membrane filtration (HCMF) and hybrid polymeric membrane filtration (HPMF) were compared in a hypothetical implementation in an existing WWTP in Switzerland, based on results from a pilot-scale implementation [26]. Previous research by Löwenberg et al. [23] suggest that 99% of benzotriazole, 93% of carbamazepine, 76% of diclofenac, 77% of mecoprop and 54% of sulfamethoxazole would be removed from secondary treated wastewater at a PAC dosage of 15 mg L⁻¹. Both HPMF and HCMF would reduce ECs in effluents with small overall environmental differences between the two systems. Compared to the current WWTP operation and taking into account background processes from PAC production and electricity, human and ecotoxicity potential would be reduced by ca. -45% and -30% to -35% per unit treated wastewater, respectively. Carbon emissions related mainly to PAC production are the main consideration regarding environmental trade-offs, with ca. 0.2 kg $CO_{2eq.}$ m⁻³ wastewater treated ([26], calculated mainly from Bayer et al. [27]) at a dosing of 15 mg L^{-1} as in this pilot-scale study. Given that the carbon balance in the studied WWTP is almost neutral due to energy recovery from sludge and the Swiss electricity mix, this is relevant. Uncertainties are substantial in this case, as PAC producers do not communicate the specific ingredients and origins of their products. In addition, uncertainties about ceramic membrane lifetime under real operational conditions for advance wastewater treatment make both environmental and economic considerations difficult at the current stage. Assuming a lifetime of seven years for polymeric membranes and 12 years for ceramic membranes, live cycle cost per unit treated water are ca. 4% higher for ceramic compared to polymeric membrane filtration.

Online stakeholder interviews [10] showed that success factors for implementation of the technology consist mainly of very high expertise levels of the involved stakeholders (especially of the research partner), a highly motivated client (or launching customer) that is open to innovative ideas and willing to take part in experimenting, and good project planning. Cooperation among involved stakeholders went mostly smooth and efficient due to clear communication on all levels, shared goals and ambitions, and knowledge about each other's core strengths and capacities. The major challenge that emerged from the surveys is about the need for a shared sense of responsibility for and ownership of the whole project, instead of only focusing on the direct responsibilities of the involved individuals.

3.2.4 USPs and recommendations for impact

Based on the preceding considerations, the following environmental and economic USPs can be derived (adapted from Gross et al. [11]):

- Over the lifecycle of a WWTP hybrid membrane filtration with ceramic membranes (HCMF) is about as expensive as with polymeric membranes (HPMF), due to an expected longer lifetime of ceramic membranes.
- The ecological performance of HCMF is slightly better than HPMF, due to a smaller membrane area required and related lower energy demand for aeration used for cleaning the membranes.
- PAC in combination with ceramic or polymeric membrane filtration provides a highly effective broadband elimination of ECs with complete particle retention, disinfection properties and no byproducts. On the other hand, PAC energy-intensive production also causes noteworthy environmental impacts especially with regard to the global warming potential.

The following main recommendations for impact can be derived (adapted from Gross et al. [17]):

- Administration and policy level: Support environmentally friendlier PAC production and transparency on raw products, which commonly include anthracite, bituminous or lignite coal, peat, wood and/or coconut shells [27]. Promote research on renewable raw materials such as agricultural and food industry by-products such as nut shells or fruit stones [28,29].
- Scientific community and technology developers: Search for ways to improve the environmental
 profile of PAC utilization along its life cycle from sourcing to deposition and/or reactivation, where
 possible. Strategies to minimize dosing include utilization of PAC's maximum adsorption capacity and
 potential optimization via ANCS and/or bioassay based monitoring. Reduce aeration by advanced
 aeration techniques, e.g. by intermittent aeration or by utilization of non-aerated pressurized
 membrane systems.

3.3 Advanced Oxidation Techniques for waste and drinking water treatment

Oxidation and advanced oxidation techniques can be used to degrade a broad range of ECs in both drinking and wastewater. Currently there are few full-scale operational plants to demonstrate the removal of ECs, apart from ozonation and advanced oxidation with UV/H_2O_2 in drinking water treatment. As part of the DEMEAU project, demonstration studies applying ozonation were conducted for both drinking and wastewater. In addition, new types of UV/H_2O_2 reactors were applied.

3.3.1 Readiness to tackle emerging contaminants

Ozone is a powerful oxidizing agent that effectively degrades many organic compounds; particularly those which contain electron rich moieties like activated aromatic compounds or tertiary amines [30]. The addition of hydrogen peroxide leads to a faster decomposition of ozone to hydroxyl radicals, which are able to abate more persistent ECs in water with low natural organic matter (NOM). Advanced wastewater treatment with ozonation can, depending on specific pre-treated wastewater properties, eliminate more than 80% of ECs therein [31].

For water containing high amounts of bromide and low concentrations of NOM, the use of ozone is risky, as bromate may be formed as a suspected carcinogenic by-product [32,33]. Ozonation is therefore problematic in drinking water production with high levels of bromide, e.g. in most Dutch surface waters. For the mitigation of bromate formation, ozone/ H_2O_2 may be applied, or UV/ H_2O_2 , where no bromate is formed. UV/ H_2O_2 comprises the photolysis of organic compounds by absorption of the UV irradiation and the photolysis of H_2O_2 resulting in the formation of hydroxyl radicals that oxidize a broad variety of contaminants.

Both oxidative technologies, ozonation with and without addition of H_2O_2 , and UV/H_2O_2 , were studied within the DEMEAU framework. The results showed that ozone, ozone/ H_2O_2 , and UV/H_2O_2 can effectively convert a broad range of ECs. A decision basis for the implementation of oxidation technologies to eliminate ECs was developed, where different parameters that influence the efficiency of oxidation and the formation of by-products are discussed [34]. The tool helps to decide if oxidative treatment of wastewater is recommended and under which circumstances the treatment of drinking water with ozone, O3/H2O2, or UV/H2O2 may be the treatment of choice.

3.3.2 Success stories

Ozonation for wastewater treatment

The WWTP Neugut in Dübendorf (CH), assessed in case study OT1, is the first wastewater treatment plant to comply with the new Swiss Water Protection Act that requires the implementation of an additional treatment step at selected plants to abate ECs. Within DEMEAU, the effectiveness of the ozonation technology regarding chemical and ecotoxicological water quality parameters at WWTP Neugut was investigated. Different doses of ozone were applied to determine the optimal conditions for removing selected ECs by more than 80% on average as required by the regulation [35]. Investigations on the behavior of certain substances under ozonation and elucidation of transformation products improved the understanding of the mechanism of ozone reactions with ECs [36]. Two toxic by-products, bromate and N-nitrosodimethylamine (NDMA), were analyzed according to a modular testing framework to evaluate the treatability of wastewater with ozone proposed by [37], and were below the drinking water standards of the WHO after dilution in the receiving water body. Experimentation with different bioassays revealed that the wastewater treatment with ozone resulted in significantly reduced ecotoxicological effects compared to conventionally treated wastewater. Biological post-treatment contributed to further decreases and is generally recommended after ozonation as it eliminates easily degradable and potentially toxic ozonation reaction products like NDMA.

Ozonation for drinking water treatment

The efficiency of conventional ozonation and the advanced oxidation process O_3/H_2O_2 under various conditions was studied in a pilot plant for the treatment of a surface water in Switzerland [38]. Compounds reacting fast with ozone were well abated (>90%) even for the lowest ozone dose of 0.5 mg/L, but compounds only reacting with hydroxyl radicals showed much lower abatement. Generally, the abatement efficiency increased with higher ozone doses, higher pH and lower bromide concentrations. H_2O_2 addition accelerated the ozone conversion to hydroxyl radicals, which enabled a faster abatement of ozone-resistant micropollutants. It was demonstrated that the formation of bromate could be mitigated by H_2O_2 addition. Post-treatment by granular activated carbon (GAC) filtration enabled the reduction of microcontaminants and selected ozonation transformation products, but no changes in bromate were observed.

UV/H₂O₂ for drinking water production

Currently, the UV/H_2O_2 process is applied in reactors that have been optimized for UV disinfection with lower UV doses (20-70 mJ/cm²) than are needed for advanced oxidation to abate ECs (about 500 mJ/cm²). Both reaction kinetics (of photolysis and oxidation) and flow through the UV reactor were modeled, and the results were used to design new UV reactors, optimized for advanced oxidation applications. These improved oxidation reactors were tested under different conditions at two drinking water utilities in the Netherlands, at Dunea and at the utility WML in Limburg. They showed a 70 - >80% decrease in energy demand, which was particularly important given that the high electricity requirements of traditional UV reactors are considered a major obstacle to broader application of the technology [39]. Despite higher costs, an UV/H₂O₂ treatment may be considered in case of high bromide concentration in the raw water.

The results also showed that the water quality of influent water directly affects the overall efficiency of the technology. Treatment of water with a high UV-transmission (UV-T >85%) is more efficient and less energy demanding, whereas influent water with a lower UV-T (\pm 75%) shows a sharp increase in energy costs. Pre-treatment with O₃/H₂O₂, or activated carbon of water with a lower UV-T, resulted in higher effectiveness and a lower energy demand by 30-70%, depending on the ECs being removed. [39]

3.3.3 Environmental, economic and societal considerations of AOP case studies

Both direct and indirect oxidation via ozone (O_3) and hydroxyl radicals (OH·) can increase the degradability of many ECs for subsequent biological degradation [40], e.g. in a sand filter. While oxidative processes have a long history in water treatment e.g. for disinfection and odor and taste elimination, targeting ECs is more recent. Applications include the advanced treatment of wastewater, drinking water, and integration in MAR schemes (*c.f.* DEMEAU case study MAR1).

The OT1 case study at WWTP Neugut was conducted on the newly built full-scale ozonation at the existing WWTP at the time of study (2015). Considering reduced emissions to local water resources based on the monitored 11 substances as well as background processes from installations and operations associated with ozonation – at 0.62 g O_3 g⁻¹ DOC (dissolved organic carbon) or ca. 3 mg O_3 L⁻¹_{wastewater} – , application of this technology resulted in no overall changes (including emissions from background processes) with regard to freshwater ecotoxicity, minor changes regarding human cancer related toxicity (+4%) and –44% reduction in human non-cancer toxicity. If the impacts of water emissions are extrapolated to a full load that exceeds by far the 11 monitored substances, this impact reduction is magnified as -39% and -51% respectively for freshwater ecotoxicity and human toxicity non-cancer effect. The carbon footprint is mainly due to the main plant infrastructure, operation and electricity. The energy consumption for local ozone generation from oxygen, which in the Swiss case study is electricity generated from hydro- and nuclear power, results in a low additional carbon footprint of ca. +8%. Already existing sand filters and favorable positioning of the ozone reactor resulted in a low investment cost in the Swiss case study. Overall, ozonation added ca. +3% to life cycle costs compared to the WWTP without ozonation.

Online stakeholder interviews [10] uncovered perceived drivers of successful implementation. Those were: positive past collaborative experiences among the involved stakeholders (resulting in trust and knowledge on 'which buttons to push' in challenging circumstances), involvement of authorities (leading to funding and legislative support), the innovative and open culture at the launching water utility, and the motivation of operating staff to acquire the required skills. One of the critical success factors that came up from the surveys was the process of combining expertise from various stakeholders (with knowledge on ecology, technology, decision-making, feasibility, etc.) to find pragmatic solutions that all stakeholders agree on.

3.3.4 USPs and recommendations for impact

Based on the preceding considerations, the following environmental and economic USPs can be derived (adapted from Gross et al. [11]):

- Ozonation with subsequent biological sand filtration as last treatment step in wastewater treatment provides an effective broadband elimination of emerging contaminants. The magnitude of environmental trade-offs depends mainly on the source of electric energy used.
- The additional ozonation step can be installed and operated economically in existing WWTPs, especially
 if the conditions at the WWTP are favorable. Such cost-saving conditions include a design allowing an
 operation without additional pumping requirements and the existence of a subsequent biological
 filtration step.

The following main recommendations for impact can be derived (adapted from Gross et al. [17]):

- Administration and policy level: Foster integrative decision making involving scientist, administration, public and NGOs, which was mentioned by various key stakeholders in case study OT1 as key driver for implementation. Furthermore, a pro-active and transparent communication from an early stage was mentioned to promote momentum and trust in the process. Decide on a dependable list of target compounds, as technology selection is also depended on which compounds should be removed.
- Scientific community and technology developers: Further explore effects of ECs and oxidation byproducts to assure that relevant target compounds are identified and effective measures are taken. Continue efforts to improve monitoring of EC effects (e.g. using bioassays) and identified target compounds.

3.4 In vitro-bioassays for water quality monitoring

Efficient monitoring and adequate treatment of water sources for potentially harmful ECs are essential for avoiding direct health risks to drinking water consumers and the environment. Current monitoring strategies mostly rely on chemical analysis, which is generally accepted in regulatory frameworks. The scope of chemical analysis is, however, restricted to the quantification/analysis of specific, targeted compounds. *In vitro* bioassays are (bio)analytical techniques that can provide either a stand-alone or a complementary monitoring technology in addition to single-substance chemical analysis. They identify the biological activity of a single substance or the combined effect of substances (known and unknown ECs) present in an environmental sample. Bioassays offer the potential to increase the rigor and scope of current water quality monitoring and provide the opportunity for measuring (eco)toxicological effects of chemicals with a common mode of action, regardless of their structure, concentration and identity [41,42]. The most relevant toxicological effects of concern considered for water quality monitoring include: acute toxicity, endocrine disruption, genotoxicity, xenobiotic metabolism, or oxidative stress [44]. When combined with chemical analytics a powerful and cost effective system for water quality assessment can be designed.

However, these technological innovations also face barriers including current legislation, which need to be revised in order to allow the use of these new technologies. Therefore, consensus building, standardisation and demonstration studies at launching facilities are essential to this process.

3.4.1 Success stories

Currently, a large variety of test systems are practically available. It is important to note, that the selection of bioassay(s) used for monitoring predetermines the type of toxicants eventually identified. As one of the first steps towards the broader implementation of *in vitro* bioassays in the water sector, DEMEAU has selected an optimal panel of bioassays suitable to assess the effect of ECs in aquatic environments with a focus on human health safety (drinking water) and modes of action relevant for the WFD compounds [45]. In total, more than a hundred bioassays were evaluated for the earlier defined toxic pathways [44]. Among others, the following criteria were considered to select assays for the panel: availability of a standardized protocol, performance characteristics, costs, support and service availability, applicability to complex sample matrices.

In order to perform rapid and cost-effective water quality assessment, the capacity of the assays for high throughput screening (HTS) is important. Within DEMEAU, HTS of a large selection of chemicals (n > 60; including the entire list of WFD chemicals, and other relevant routinely monitored chemicals by the DEMEAU partners) was conducted with an extended bioassay panel using a certain type of *in vitro* bioassays with human cell lines (CALUX[®] technology). These bioassays have been shown to be powerful tools to assess toxicity of a much wider range of chemicals than currently monitored [46,47,48,49,50,51]. The HTS revealed that some 70% of WFD and other routinely monitored chemicals were biologically active. The reason for this is that several chemicals are monitored because of their ubiquitous presence in the aquatic

environment rather than their inherent toxicity. The study suggested that the originally proposed panel of assays may still need some expansion. Furthermore, hazard identification and risk assessment of hugely complex chemical mixtures as being present in environmental water samples should not rely solely on lists of prioritized chemicals, which warrants inclusion of biological methods, which now yield reliable and quantitative results.

In addition to screening of the presence of target substances, bioassays were applied for evaluation of the EC removal efficiencies of the DEMEAU technologies. Screening of various types of MAR sources (groundwater, surface water and WWTP effluent) revealed the importance of endocrine, oxidative stress and photosynthesis inhibition pathways. Clear differences between clean and polluted sites could be detected [52,53]. Such studies can help improving the performance of the MAR sites by identifying those sites where further actions are needed to reveal the identity and sources of the compounds causing the measured effects.

For both, hybrid ceramic membrane filtration (HCMF) and advanced oxidation techniques (OT), *in vitro* bioassays can be used to evaluate their proper functioning. This is done by screening the total toxic potency of the untreated and released product (i.e. drinking water or wastewater effluents). Screening of a HCMF (PAC-UF) system for the removal of ECs from municipal wastewater showed substantial reduction of hormonal effects, bacteria luminescence inhibition and photosynthesis inhibition in algae as induced by the spiked chemicals. Wastewater treatment with ozone resulted in significantly reduced activity in the majority of bioassays as compared to effects measured in the conventionally (biologically) treated wastewater. Increased activities after ozonation occurred in a few assays, which could be removed by suitable post-treatments.

For any analytical approach, it is important to set realistic thresholds to be able to perform an assessment of potential risks for human health or the environment. Thus, an important step in the regulatory acceptance of bioassay data is the creation of so called "trigger values" (i.e. thresholds or cut off values) defining a level above which human health or ecosystem risks cannot be waived [43,54]. Such trigger values have also been suggested and applied in frame of DEMEAU.

3.4.2 Environmental, economic and societal considerations of *in vitro* bioassay case studies

Invitro bioassays have emerged as a promising tool to assess potentially harmful effects of mixtures of contaminants in water, even if the identity of the compounds present are not known. *In vitro* bioassays could complement chemical and physical monitoring, providing opportunities for integration into existing monitoring strategies. Key application areas with regard to ECs would be the monitoring of potential toxicological risks, including the quantification of EC reduction measures such as technologies studied in DEMEAU.

No detailed environmental and economic assessments were performed for *in vitro* bioassays in DEMEAU. Additional energy and other resources for water quality monitoring are most likely not significant in comparison to resources needed for water treatment. *In vitro* bioassays could also be used as a screening tool, e.g. in case study BA1 (see Table 1), to partially replace chemical analysis by LC-MS/MS, which in that case is only used when bioassays indicate some effects.

Online stakeholder interviews [10] resulted in an overview of drivers for implementation of in vitro bioassays. One such driver is the compatibility of bioassay monitoring with existing practices; they would be complementing existing chemical analysis techniques in very valuable ways and would not disturb the present monitoring process much. With regard to the implementation context, due to the innovative character of in vitro bioassays, the broad possibilities are still unknown to many potential end users in the water sector. Next to that, regulations do not yet cover thresholds on compound mixture effects; without regulations or standards, high levels of intrinsic motivation (to learn, gain awareness) are required at the launching water utilities for successful implementation. The relation between demand (willingness to

invest) and capacity/opportunity to further develop the technology is also mentioned as an important factor: when the demand is low, investments stay behind, being a barrier to further development,. This, in turn, does not help to raise the demand and/or levels of knowledge with regard to this technology. Progress depends thus on knowledgeable research institutes and (public) funding programmes.

3.4.3 Unique selling points and recommendations for impact

Based on the preceding considerations, the following environmental and economic unique selling points USPs can be derived (adapted from Gross et al. [11]):

- Effect-based *in vitro* bioassays can give additional information in water quality monitoring towards unknown or emerging compounds with potential adverse effects.
- *In vitro* bioassays for groups of compounds with similar mode of action can provide a cost-effective way to screen water quality by substituting a significant number of chemical analyses.
- Effect-based methods provide robust monitoring tools to better benchmark and better ensure ecosystem and human health risk regarding trigger levels designed for a more proactive water treatment management.

The following main recommendations for impact can be derived (adapted from Gross et al. [17]):

- Administration and policy level: Develop standards and guidelines for applications, taking advantage of the fact that *in vitro* bioassays detect presence of potentially hazardous ECs even without specific knowledge of the compounds.
- Scientific community and technology developers: Continue and intensify efforts to convey the concept of toxicological safety regarding ECs to non-expert stakeholders, potentially lowering barriers to communicate results from *in vitro* bioassay screenings.

3.5 Further technologies explored in DEMEAU

Artificial Neural Networks in combination with a Genetic Algorithm (GA) optimizer have been successfully applied to water treatment technologies as part of DEMEAU. The combination is called Automatic Neural Net Control System (ANCS). ANCS is primarily a computer-based system searching for process optimization that is fed with input signals from a technical process, such as sensor-data of a drinking water plant. In this way, the ANCS uses input information to determine the optimal performance of the process at hand. In the case of a drinking water plant, such a process could be a membrane filtration process, where the system aims to optimize its target parameters. Such parameters include permeability, energy consumption, or cost efficiency.

3.6 Comparison / Synergies

DEMEAU showcased the diverse and also synergetic application areas of the described technologies and their potential for removing or monitoring the removal of ECs from drinking and wastewater.

LCAs and LCCs demonstrated that interventions to remove emerging contaminants can have significant benefits on the local water quality, but are also accompanied by additional environmental impacts and costs. This is mainly due to required installations and resource demands such as labor, electricity and/or chemicals. Most environmental impacts are associated with the production of chemicals and energy needed and thus do not occur at the water treatment plant directly. Co-benefits, such as a minimization of additional impacts and pollution through ECs at the sources, should be considered where feasible, to maximize the removal of ECs. The layout and operation of EC removing technologies should optimize the resource demand. Both ANCS and *in vitro* bioassays provide synergetic options regarding this goal. ANCS, in particular, can be used to optimize the operation of drinking water treatment plants and electricity and may help improve environmental and economic footprints by cutting-down on chemicals and electricity

demand and possibly reduced installation needs. *In vitro* bioassays have strong synergies with all EC removal technologies and can be applied to monitor their effectiveness, and highlight local (eco)toxicological risks. They have already been applied to assess the feasibility of ozonation for wastewater treatment in combination with other parameters [37] as well as for a performance review of various advanced treatment techniques (e.g. [55]).

Various synergies between the ECs removal technologies studied in DEMEAU have been highlighted and could improve future implementations. Applied in combination with ozonation or UV treatment, MAR can be used to remove by-products from the water. The capacity of MAR systems to remove ECs or by-products depends on site specific conditions.

Depending on the use, pre-treatment of the source water and post-treatment of the abstracted water is possible using AOTs or H(C)MF [56]. Furthermore, treatment lines combining oxidative processes with H(C)MF have been used in drinking water treatment to take advantage of the individual strengths of the two technologies regarding removal of specific ECs. Such synergies could also be an option for advanced wastewater treatment.

4 Barriers and how to foster implementation of innovative water technologies

The selection of treatment technologies to abate ECs depends on a number of factors including ease of integration into existing infrastructure and operations, available space, composition of the (pre-treated) wastewater, ECs present in the source water and possible future specific regulations for target compounds. During the project, several barriers to the implementation of innovative technologies were uncovered. Regulation and political incentives are currently insufficient. Therefore, it could be useful to integrate regulators and policy makers closer into the project cycle.

With regards to regulation, *in vitro* bioassays and MAR are particularly affected. In contrast to *in vivo* assays, which have already been applied for these purposes since decades, most *in vitro* bioassays are currently not included in the standardized monitoring routines to assess ecotoxicological effects and mixture toxicity, potentially due to the lack of knowledge of the technology on the part of the authorities. This lack of regulatory demand hampers interest of potential end-users in water utilities and the allocation of funds for validating the technology. MAR application is site-specific and requires individual assessment. The difficulty in generalization across cases makes a general policy or regulation challenging. However, early involvement of the local authorities has proven as an effective method for overcoming this barrier. So far, cooperation between authorities, water utilities and scientists is poorly developed and therefore no relevant changes in regulation are likely to be achieved.

Here, political incentives have the power to create enabling and less risky environments. The currently existing lack thereof poses a barrier to the implementation of innovative technologies, particularly with regard to the implementation of the advanced treatment in water utilities or the requirement of energy efficient water treatment. With the funding of the project, several innovations could be developed further and it was therefore understood as an economic incentive. On the other hand, the lack of financial resources led to ceasing projects e.g. in Spain (MAR).

Barriers to implementation stem from a variety of factors, including lack of knowledge and awareness on the side of authorities and the public [10]. This latter barrier can be addressed through targeted communication, knowledge transfer and cooperation amongst sectors. At the same time, knowledge gained needs to be transferred into understandable information to the relevant stakeholders while also connecting to stakeholders' worldviews. Practitioners also indicated that the decision is based on positive experiences with a specific technology at comparable sites. Effective communication to the public can also help to overcome a negative but uninformed public opinion, in addition to good cooperation and previous collaborative experience.

5 Conclusions and outlook

Amongst the investigated technologies that have the ability to remove ECs MAR, HCMF (in combination with PAC treatment), ozonation and UV/H_2O_2 treatment are promising candidates for many tasks in future water treatment schemes.

MAR by natural infiltration ponds are a low-cost and low-energy option for groundwater recharge and can be implemented. In addition, these ponds can be upgraded or combined with other process steps (e.g. pre-treatment with advanced oxidation processes) to enhance their capacity for removal of ECs.

HCMF can be applied to a diversity of water sources to produce high quality water, also removing ECs. Full scale drinking water treatment that uses HCMF is already in operation, both at PWN, as a pre-treatment for UV/H_2O_2 , and at Vitens, to reuse rinse water of sand filters (both in the Netherlands). HCMF in combination with PAC, ozonation or ion exchange can also be used in treatment of wastewater, offering opportunities for future application.

Ozonation requires less energy than UV/H_2O_2 processes and can only be applied to a limited extend if bromide is present. It is one of the most economic treatment options to abate ECs and is implemented successfully in full-scale drinking water treatment and, in recent years, for wastewater treatment. However, in some countries such as the Netherlands, surface water contains relatively high bromide concentrations, which leads to the formation of bromate, a suspected carcinogen,, upon reaction with ozone. In such a case, UV/H_2O_2 treatment is considered a useful alternative, despite the fact that energy requirements are substantially higher compared to treatment with ozone or O_3/H_2O_2 . The relatively high energy demand of **UV processes** poses a barrier to implementation of this technique, which can significantly be improved with the newly designed reactors in DEMEAU and possibilities for pre-treatment of the water. As a result, full-scale application of **UV/H_2O_2** processes can be the treatment of choice at locations with high bromide level.

At the same time, process optimization is an ongoing task for the water treatment sector. **ANCS** can be applied to a diverse range of systems and to systems of all sizes. The size of the treatment system is directly related to the economic viability of the plant, as time is an important factor when considering investment returns. In particular ANCS are well applicable to membrane filtration processes due to their highly reproducible behavior. As such, scaling up the results from a pilot plant to the full scale operating plant does not present any major problems. **ANCS** has been found to increase environmental efficiency.

On the monitoring side, *in vitro* bioassays have the ability to investigate water quality based on the specific toxic activity of the contaminants that are present, rather than their specific structures or identity. Such bioassays present a wide scope of water quality monitoring from general toxicity tests to tests assessing very specific biological activities. *In vitro* bioassays are capable of measuring the mixture effect of (un)known compounds with a similar mode of action, and therefore can act as a complementary tool to chemical-analytical techniques. However, to be able to perform an assessment of potential risks for human health or the environment, the derivation of a threshold level (trigger value, bioassay guideline value) is a crucial step towards regulatory acceptance and practical application (e.g. [43,54,57,58]).

During the project, it could be shown that the technologies can reach an even higher effectiveness concerning ECs treatment when applied in combination and further optimized compared to a stand-alone application. In an ideal case, *in vitro* bioassays could be applied for testing treated wastewater for specific activities (e.g. receptor activation) with regard to ECs and thus, indicate potential (eco)toxicity. When bioassay responses are observed in treated waste water, the water should be treated additionally with the advanced treatment technology, i.e. the so-called fourth treatment step. It is suggested that at least the effectiveness of the advanced treatments should be routinely monitored with *in vitro* bioassays. Innovative, but more resource demanding technologies such as HCMF in combination with PAC would only be used when this treatment step is legally binding and also cost-efficient. ANCS could also be applied along the process, to further optimize the technologies. Finally, water after ozonation could be further filtered and infiltrated using MAR. In such a way, the additional treatment would enhance the removal of ECs and potential by-products in case ozonation is applied.

When it comes to advancing the uptake of innovative practices and technologies to clean waste and drinking water, no universal solution is available as the underlying reasons for the slow technology uptake are manifold. A package of updated regulations, laboratory research, site demonstrations, business case evaluation and targeted information of all relevant stakeholders can trigger a shift in the water sector. Reaching the aim of full-scale application of innovative processes to tackle ECs in waste and drinking water will help to improve sustainable water resource management and secure water quality in the future. Innovation takes time; therefore, this aim is not yet achievable. However, EU funded research projects such as DEMEAU contribute to the road towards this goal. Research on the technologies mentioned in this article and carried out within DEMEAU advanced the knowledge basis created by previous EU research projects. Communication is crucial for technology uptake. The results gained through LCA and LCC of the technologies are powerful means to present technical information to policy makers and the public. The demonstration of the technologies brings the water sector closer to the goal. In a subsequent step, the potential of the synergies of these technologies needs to be fully exploited, as these technologies should not only be seen as stand-alone approaches.

Acknowledgments: The DEMEAU project was supported by the European Union Seventh Framework under grant agreement no. 308339.

6 References

- [1] NORMAN Network, Emerging substances, NORMAN. <u>http://www.norman-network.net/?q=node/19</u>, 2015 (accessed 12 March 2018).
- [2] J. Corcoran, M.J. Winter, C.R. Tyler, Pharmaceuticals in the aquatic environment: A critical review of the evidence for health effects in fish, Critical Reviews in Toxicology 40 (2010) 287– 304.
- [3] M. Wagner, C. Kienle, E.L.M. Vermeissen, J. Oehlmann, Endocrine disruption and in vitro ecotoxicology: Recent advances and approaches, in: G. Reifferscheid, S. Buchinger (Eds.), In Vitro Environmental Toxicology – Concepts, Application and Assessment, Advances in Biochemical Engineeering/Biotechnology, vol. 157, Springer, Cham, 2017.
- [4] N. Chevre, S. Erkman, Alerte aux micropolluants, second ed., PPUR, Lausanne, 2017.
- [5] EU, DIRECTIVE 2013/39/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy, in: Union EPaCotE (Ed.), L226/1, Off. J. Eur. Union, 2013.
- [6] M. Schriks, M.B. Heringa, M.M.E van der Kooi, P. de Voogt, A.P. van Wezel, Toxicological relevance of emerging contaminants for drinking water quality, Water Res. 44 (2010) 461– 476.
- [7] C. Moschet, C. Götz, P. Longrée, J. Hollender, H. Singer, Multi-level approach for the integrated assessment of polar organic micropollutants in an international lake catchment: The example of lake Constance, Env. Sci. Tech. 47 (2013) 7028-7036.
- [8] Swiss Federal Council. Mikroverunreinigungen: Bundesrat verabschiedet Botschaft zur Finanzierung.

https://www.admin.ch/gov/de/start/dokumentation/medienmitteilungen.msg-id-49455.html, 2013 (accessed 12 March 2018).

- [9] R.I.L. Eggen, J. Hollender, A. Joss, M. Schärer, C. Stamm, Reducing the Discharge of Micropollutants in the Aquatic Environment: The Benefits of Upgrading Wastewater Treatment Plants. *Env. Sci. & Tech.* 48(14) (2014) 7683-7689.
- [10]M. Pieron, M. van der Zouwen, Drivers and barriers for successful implementation of innovative DEMEAU technologies, Deliverable 52.1 from the DEMEAU project (FP 7 framework), 2014. <u>http://demeau-fp7.eu/content/d521</u> (accessed 12 March 2018).
- [11] T. Gross, A. Kounina, C. Oberschelp, C. Remy, K. Wencki, C. Hugi, Unique selling propositions of promising technologies to address emerging pollutants in water and wastewater. Deliverable of the DEMEAU project (FP 7 framework), 2015. <u>http://demeaufp7.eu/content/d511</u> (accessed 12 March 2018).
- [12]C. Remy, K. Wencki, M. Pieron, A. Kounina, C. Hugi and T. Gross, Final guidelines for sustainability assessment of water technologies (D51.2), Deliverable of the DEMEAU project (FP 7 framework), 2015. http://demeau-fp7.eu/content/d512 (accessed 12 March 2018).

- [13] R.K. Rosenbaum, T.M. Bachmann, L.S. Gold, M.A.J. Huijbregts, O. Jolliet, R. Juraske, A. Koehler, H.F. Larsen, M. MacLeod, M. Margni, T.E. McKone, J. Payet, M. Schuhmacher, D. van de Meent and M.Z. Hauschild, USEtox the UNEP-SETAC toxicity model: recommended characterization factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment, Int. J. LCA 13(7) (2008) 532-546.
- [14] C. Sprenger, N. Hartog, M. Hernández-García, E. Vilanova, G. Grützmacher, F. Scheibler, S. Hannappel, Inventory of Managed Aquifer Recharge sites in Europe – historical development, current situation and perspectives, Hydrogeo. J. 25 (2017) 1909–1922.
- [15] E. Vilanova, M. Miret, J. Molinero, Decision trees for MAR impact evaluation, Deliverable 12.1 of the DEMEAU project (FP 7 framework), 2013. <u>http://demeau-fp7.eu/content/d121</u> (accessed 12 March 2018).
- [16] P. Dillon, P. Pavelic, D. Page, H. Beringen, J. Ward, Managed aquifer recharge: an introduction, National Water Commission, Canberra, 2009.
- [17] ENSAT, Enhancement of Soil Aquifer Treatment to Improve the Quality of Recharge Water in the Llobregat River Delta Aquifer, Layman's report, CETaqua, Barcelona, 2012. <u>https://acaweb.gencat.cat/aca/documents/ca/actuacions_rdi/Fitxa_ENSAT.pdf</u> (accessed 12 March 2018).
- [18] T. Gross, A. Kounina, M. Pieron, C. Remy, K. Wencki, C. Hugi, Recommendations for impact: Promising technologies to address emerging pollutants in water and wastewater. Deliverable 52.2 of the DEMEAU project (FP 7 framework), 2015. <u>http://demeau-fp7.eu/content/d522</u> (accessed 12 March 2018).
- [19] T. Van den Hoven, TECHNEAU Safe drinking water from source to tap: State of the art and perspectives, IWA Publishing, London, 2009 (ISBN: 9781843392750).
- [20] S.H. Park, Y.G. Park, J.L. Lim, S. Kim, Evaluation of ceramic membrane applications for water treatment plants with a life cycle cost analysis, Desalin. Water Treat. 54 (2015) 973–979.
- [21] G. Galjaard, J. Clement, A. Wui Seng,L. Mong Hoo, 2012. Ceramac[®]-19 demonstration plant ceramic microfiltration at Choa Chu Kang Waterworks. Water Practice Tech., 7(4) (2012) wpt2012087. doi: 10.2166/wpt.2012.087
- [22] J. Margot, C. Kienle, A. Magnet, M. Weil, L. Rossi, L.F. de Alencastro, C. Abegglen, D. Thonney, N. Chèvre, M. Schärer, D.A. Barry, Treatment of micropollutants in municipal wastewater: ozone or powdered activated carbon? Sci. Total Env. 461-462 (2013) 480–498.
- [23] J. Löwenberg, A. Zenker, M. Baggenstos, G. Koch, C. Kazner, T. Wintgens, Comparison of two PAC/UF processes for the removal of micropollutants from wastewater treatment plant effluent: Process performance and removal efficiency, Water Res. 56(0) (2014) 26–36.
- [24] S.G. Lehman, L. Liu, Application of ceramic membranes with pre-ozonation for treatment of secondary wastewater effluent, Water Res. 43 (2009) 2020–2028.
- [25] A. Ahmad, W. Siegers, J. Svojitka, J. Löwenberg (2015), D22.1 Demonstration of Hybrid Ceramic Membrane Filtration for the Removal of Organic Micropollutants from Municipal Wastewater Treatment Plant Effluent, Deliverable of the DEMEAU project (FP 7 framework). <u>http://demeau-fp7.eu/system/files/results/D22.1.pdf</u> (accessed 12 March 2018).

- [26] W. Oberschelp, Sustainability assessment of micropollutant sequestration from pre-treated wastewater via hybrid ceramic membrane processing, master thesis, RWTH, Aachen, 2015.
- [27] P. Bayer, E. Heuer, U. Karl, M. Finkel, Economical and ecological comparison of granular activated carbon (GAC) adsorber refill strategies. Water Res. 39 (2005) 1719–28.
- [28]A. Aygün, S. Yenisoy-Karakaş, I. Duman, Production of granular activated carbon from fruit stones and nutshells and evaluation of their physical, chemical and adsorption properties, Microporous Mesoporous Mater 66 (2003) 189–195.
- [29] D. Angin, Production and characterization of activated carbon from sour cherry stones by zinc chloride, Fuel 115 (2014) 804–811.
- [30] C. von Sonntag, U. von Gunten, Chemistry of ozone in water and wastewater treatment, IWA Publishing, London, 2012.
- [31] J. Hollender, S.G. Zimmermann, S. Koepke, M. Krauss, C.S. McArdell, C. Ort, H. Singer, U. von GuntenH. Siegrist, Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. Env. Sci. Tech. 43 (2009) 7862–7869. doi: 10.1021/es9014629
- [32] E.C. Wert, F.L. Rosario-Ortiz, D.D. Drury, S.A. Snyder, Formation of oxidation byproducts from ozonation of wastewater, Water Res. 41 (2007) 1481–1490.
- [33] Hollender J, Zimmermann SG, Koepke S, Krauss M, McArdell CS, Ort C, et al. Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. Environ Sci Technol 2009;43:7862–9
- [34] C.S. McArdell, M. Bourgin, U. von Gunten, J. Hollender, C. Kienle, R. Hofman-Caris, D32.3 Decision basis for implementation of oxidation technologies, Deliverable 32.2 of the Demeau project (FP 7 framework), 2015. <u>http://demeau-fp7.eu/content/d323</u> (accessed 12 March 2018).
- [35] M. Bourgin, B. Beck, M. Boehler, E. Borowska, J. Fleiner, E. Salhi, R. Teichler, U. von Gunten, H. Siegrist, C.S. McArdell, Evaluation of a full-scale wastewater treatment plant upgraded with ozonation and biological post-treatments: Abatement of micropollutants, formation of transformation products and oxidation by-products, Water Res. 129 (2018) 486-498.
- [36] E. Borowska, M. Bourgin, J. Hollender, C. Kienle, C.S. McArdell, U. von Gunten, Oxidation of cetirizine, fexofenadine and hydrochlorothiazide during ozonation: Kinetics and transformation products, Water Res. 94 (2016) 350-362.
- [37] Y. Schindler Wildhaber, H. Mestankova, M. Schärer, K. Schirmer, E. Salhi, U. von Gunten, Novel test procedure to evaluate the treatability of wastewater with ozone, Water Res. 75 (2015) 324-335.
- [38] M. Bourgin, E. Borowska, J. Helbing, J. Hollender, H.-P. Kaiser, C. Kienle, C.S. McArdell, E. Simon, U. von Gunten, Effect of operational and water quality parameters on conventional ozonation and the advanced oxidation process O3/H2O2: Kinetics of micropollutant abatement, transformation product and bromate formation in a surface water, Water Res. (2017).

- [39] C.H.M. Hofman-Caris, D.J.H. Harmsen, A.M. van Remmen, A.H. Knol, W.L.C van Pol, B.A. Wols, Optimization of UV/H2O2 processes for the removal of organic micropollutants from drinking water; effect of reactor geometry and water pretreatment on EEO values, Wat. Sci.Techn. 17(2) (2017) 508-518.
- [40] C. Abegglen, H. Siegrist, Mikroverunreinigungen aus kommunalem Abwasser. Verfahren zur weitergehenden Elimination auf Kläranlagen. Bundesamt für Umwelt, Berne, 2012.
- [41] A. Jia, B.I. Escher, F.D.L. Leusch, J.Y.M Tang, E. Prochazka, B. Dong, E.M. Snyder, S.A. Snyder, In vitro bioassays to evaluate complex chemical mixtures in recycled water, Water Res. 80 (2015) 1-11.
- [42] A.-S. Wernersson, M. Carere, C. Maggi, P. Tusil, P. Soldan, A. James, W. Sanchez, V. Dulio, K. Broeg, G. Reifferscheid, S. Buchinger, H. Maas, E. van der Grinten, S. O'Toole, A. Ausili, et al., The European technical report on aquatic effect-based monitoring tools under the water framework directive, Env. Sci. Eur. 27(7).
- [43] B.I. Escher, S. Aït-Aïssa, P.A. Behnisch, W. Brack, F. Brion, A. Brouwer, S. Buchinger, S.E. Crawford, D. du Pasquier, T. Hamers, K. Hettwer, K. Hilscherová, H. Hollert, R. Kase, C. Kienle, A.J. Tindall, J. Tuerk, R. van der Oost, E. Vermeirssen, P.A. Neale, Effect-based trigger values for in vitro and in vivo bioassays performed on surface water extracts supporting the environmental quality standards (EQS) of the European Water Framework Directive. Sci. Total Env. 628-629 (2018) 748-765.
- [44] B.I. Escher, M. Allinson, R. Altenburg, P.A. Bain, P. Balaguer, W. Busch, J. Crago, N.D. Denslow, E. Dopp, K. Hilscherova, A.R. Humpage, A. Kumar, M. Grimaldi, B.S. Jayasinghe, B. Jarosova, et al., Benchmarking Organic Micropollutants in Wastewater, Recycled Water and Drinking Water with In Vitro Bioassays, Env. Sci. & Tech. 48(3) (2014) 1940-1956.
- [45] M. Schriks, K. Baken, E. Simon, H. Besselink, S. van der Linden, C. Kienle, B. van der Burg, (2014). Selection criteria to select in vitro bioassays for implementation and use, Deliverable 41.1 of the DEMEAU project (FP 7 framework), 2014. <u>http://demeau-fp7.eu/content/d411</u> (accessed 12 March 2018).
- [46] B. Van der Burg, S.C. Van der Linden, H.Y. Man, R. Winter, L. Jonker, B. van Vugt-Lussenburg, A. Brouwer, A panel of quantitative CALUX[®] reporter gene assays for reliable high throughput toxicity screening of chemicals and complex mixtures, in: P. Steinberg (Ed.), High throughput screening methods in toxicity testing, John Wiley and Sons, Inc. New York, pp. 519-532. ISBN 9781118065631
- [47] B. Pieterse, E. Felzel, R. Winter, B. van der Burg, A. Brouwer, PAH-CALUX, an optimized bioassay for carcinogenic hazard identification of polycyclic aromatic hydrocarbons (PAHs) as individual compounds and in complex mixtures, Env. Sci. Technol. 47 (2013) 11651-11659.
- [48] S.C. Van der Linden, A. von Bergh, B. Van Vugt-Lussenburg, L. Jonker, A. Brouwer, M. Teunis,
 C. Krul and B. Van der Burg, Development of a panel of high throughput reporter gene assays to detect genotoxicity and oxidative stress, Mutation Res. 760 (2014) 23-32.
- [49] B. Van der Burg, B. Pieterse, H. Buist, G. Lewin, S.C. Van der Linden, H.Y. Man, A.H. Piersma,E. Rorije, I. Mangelsdorf, A.P. Wolterbeek, E.D. Kroese, B.M. Van Vugt-Lussenburg, A high

throughput screening system for predicting chemically-induced endocrine disruption and sex organ deformities, Reprod. Toxicol. 55 (2015) 95-103.

- [50] B. Pieterse, I. Rijk, E. Simon, B.M. Van Vugt-Lussenburg, B.F. Fokke, M. Van der Wijk, H. Besselink, B. Van der Burg, Effect-based assessment of a persistent organic pollutants and a pesticide dump using mammalian reporter cell lines, ESPR 22 (2015) 14442-14454.
- [51] C. Brussaard, L. Peperzak, S. Beggah, L.Y. Wick, B. Wuerz, J. Weber, J.S. Arey, B. Van der Burg, A. Jonas, J. Huisman, J.R. Van der Meer, Immediate Ecotoxicological Effects of Short-Lived Marine Oil Spills Detected by Rapid on-site Biological Assays on Marine Biota, Nature Communications 7 (2016) 11206-11216.
- [52] B. de la Loma González, C. Sprenger, C. Kienle, E. Simon, G. Grützmacher, H. Besselink, M. Hernández, N. Hartog, O. Gibert, W. Seis, D11.2 Demonstration of MAR effects on groundwater resources development and application of different approaches for risk and impact assessment, Deliverable 11.2 of the DEMEAU project (FP 7 framework), 2015. http://demeau-fp7.eu/content/d112 (accessed 12 March 2018).
- [53] K. Baken, H. Besselink, A. Herbert, C. Kienle, M. Schriks, E. Simon, B. van der Burg, Report on robustness of novel water treatment technologies as determined with bioassays in a novel testing framework, DEMEAU, 2015. <u>https://www.dora.lib4ri.ch/eawag/islandora/object/eawag:14625</u> (accessed on 12 March 2018).
- [54] W. Brand, C.M. de Jongh, S.C. van der Linden, W. Mennes, L.M. Puijker, C.J. van Leeuwen, A.P. van Wezel, M. Schriks, M.B. Heringa, Trigger values for investigation of hormonal activity in drinking water and its sources using CALUX bioassays, Env. Int. 55 (2013) 109-118.
- [55] C. Kienle, R. Kase, I. Werner, Evaluation of bioassays and wastewater quality In vitro and in vivo bioassays for the performance review in the project "Strategy Micropoll", Swiss Centre for Applied Ecotoxicology Eawag-EPFL, Dübendorf, 2011.
- [56] S.K. Maeng, S.K. Sharma, K. Lekkerkerker-Teunissen, G.L. Amy, Occurrence and fate of bulk organic matter and pharmaceutically active compounds in managed aquifer recharge: A review, Water Res. 45 (2011) 3015-3033.
- [57] P.Y. Kunz, C. Kienle, M. Carere, N. Homazava, R. Kase, In vitro bioassays to screen for endocrine active pharmaceuticals in surface and wastewaters. J. Pharm. Biomed. Anal. 106 (2015) 107-115.
- [58] B.I. Escher, P.A. Neale, F.D.L. Leusch, Effect-based trigger values for in vitro bioassays: Reading across from existing water quality guideline values, Water Res. 81 (2015) 137-148.