Environmental Science & Technology

Perspective

Cite This: *Environ. Sci. Technol.* 2018, 52, 5062−5075

pubs.acs.org/est

Oxidation Processes in Water Treatment: Are We on Track?

Urs von Gunten*,[†,‡,§]

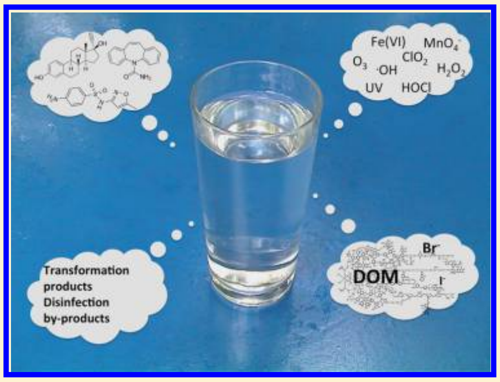
[†]Eawag, Swiss Federal Institute of Aquatic Science and Technology, Ueberlandstrasse 133, 8600 Duebendorf, Switzerland

[‡]School of Architecture, Civil and Environmental Engineering (ENAC), École Polytechnique Fédérale de Lausanne (EPFL), 1015, Lausanne, Switzerland

[§]Institute of Biogeochemistry and Pollutant Dynamics, ETH Zurich, 8092 Zurich, Switzerland

Supporting Information

ABSTRACT: Chemical oxidants have been applied in water treatment for more than a century, first as disinfectants and later to abate inorganic and organic contaminants. The challenge of oxidative abatement of organic micropollutants is the formation of transformation products with unknown (eco)toxicological consequences. Four aspects need to be considered for oxidative micropollutant abatement: (i) Reaction kinetics, controlling the efficiency of the process, (ii) mechanisms of transformation product formation, (iii) extent of formation of disinfection byproducts from the matrix, (iv) oxidation induced biological effects, resulting from transformation products and/or disinfection byproducts. It is impossible to test all the thousands of organic micropollutants in the urban water cycle experimentally to assess potential adverse outcomes of an oxidation. Rather, we need multidisciplinary and automated knowledge-based systems, which couple predictions of kinetics, transformation and disinfection byproducts and their toxicological consequences to assess the overall benefits of oxidation processes. A wide range of oxidation processes has been developed in the last decades with a recent focus on novel electricity-driven oxidation processes. To evaluate these processes, they have to be compared to established benchmark ozone- and UV-based oxidation processes by considering the energy demands, economics, the feasibilty, and the integration into future water treatment systems.



INTRODUCTION

At the beginning of the 20th century, chemical oxidants (e.g., chlorine, ozone) were introduced in municipal water treatment for disinfection in industrialized countries, which are the focus of this paper.[1,2] The combination of chemical disinfection with sand filtration led to a dramatic decrease of waterborne diseases.[1] Though, this was recognized as a major advance, the detection of the disinfection byproduct (DBP) chloroform in the mid-1970s[3,4] followed by the discovery of hundreds of other halogenated DBPs formed during chlorination and the potential link between DBPs and bladder cancer or miscarriages[5] led to substantial changes in water treatment practices. DBPs are formed from the reactions of chemical oxidants with matrix components such as the dissolved organic matter (DOM), bromide and iodide.[6−8] During ozonation, DBPs such as bromate[9] and oxygen-rich compounds such as aldehydes, ketones and carboxylic acids are formed from reactions with bromide or DOM, respectively.[10−12] In response to concerns about DBPs, other disinfectants such as chloramine[6] or chlorine dioxide[13] were applied in distribution systems. This led to significantly lower THM formation, however, other DBPs such as *N*-nitrosodimethylamine (NDMA)[14,15] or chlorite/chlorate,[16,17] respectively, were formed.

The trade-off between disinfection and DBP formation (i.e., between acute and chronic toxicity) has to be carefully evaluated.[6,8] Generally, disinfection has the highest priority and should not be compromised by the control of DBPs.[18] Nevertheless, in some countries, the use of chlorine and/or chemical disinfection in distribution systems has been entirely discontinued to avoid public health consequences and chlorine-induced taste and odor problems.[19,20] Such changes require a very rigorous approach focusing on water resources protection, natural attenuation, multibarrier treatment, and careful maintenance of distribution systems.[21]

OXIDATIVE ABATEMENT OF ORGANIC MICROPOLLUTANTS

Range of oxidation processes. Almost in parallel to chemical disinfection, oxidative control of aesthetic parameters such as iron(II) and Mn(II),[22−24] color and taste and odor[25] and other inorganic contaminants (sulfide, nitrite and ammonia) was implemented in water treatment.[26−28] Later on, oxidative treatment was applied for abatement of synthetic organic compounds (micropollutants) due to their widespread detection in drinking water sources.[5,29,30]

Received: January 30, 2018
Published: April 19, 2018

DOI: 10.1021/acs.est.8b00586

Numerous chemical oxidants have been tested alone or in combination with adsorption (activated carbon) or tight membrane processes for an optimized abatement of micropollutants (Figure 1). The tested chemical oxidation processes include ozone (O_3) and the corresponding advanced oxidation processes (AOPs), O_3/H_2O_2, O_3/UV to enhance hydroxyl radical ($^{\bullet}OH$) formation,[27,31] ultraviolet light (UV) combined with H_2O_2 to produce $^{\bullet}OH$,[32−34] permanganate,[35,36] bisulfite-activated permanganate,[37,38] the (photo)-Fenton process,[33,39] UV combined with $S_2O_8^-$ or HSO_5^- to produce sulfate radicals[40−47] and their application for in situ chemical oxidation[48,49] and ferrate(VI).[50,51] A significant effort was made over decades to develop photocatalytic water treatment.[52−55] However, some researchers were skeptical already at an early stage of investigations on TiO_2-based photocatalysis, and also today there are only limited niche applications.[56−59] Overall, in drinking water practice, most full-scale processes employ homogeneous ozone- or $^{\bullet}OH$-based processes due to their broadband action against many different classes of micropollutants,[27,60] their maturity and/or relatively low energy demand, the controllable formation of DBPs and the relatively easy implementation at full-scale (Figure 1).[2,32,61,62]

From Kinetics to Transformation Products. In early applications of oxidation processes, most studies focused on the disappearance of target compounds.[63−66] However, already during the development phase of oxidation processes, researchers recognized that typical chemical oxidant doses applied in water treatment do not yield full mineralization of most target compounds, but instead transformation products are formed.[67−70] Despite the parallel research on disinfection byproduct formation, this fact was largely neglected until the new EU drinking water regulation in 1998 applied the same standard for pesticides and transformation products/metabolites.[71] Especially in France, this came as an unpleasant surprise, because many drinking water treatment plants had been upgraded to use the O_3/H_2O_2 process in the late 1990s as a means of treating atrazine and other herbicides.[72−74] Subsequently, the O_3/H_2O_2 process was banned in France for treatment of source waters containing pesticides; today it is only approved in France for the treatment of chlorinated compounds.[75] After this caesura, more research efforts were dedicated to the identification of transformation products from common contaminants (e.g., atrazine, methyl *tert*-butyl ether, dioxane) and model compounds to provide insights into oxidative reaction mechanisms.[76−82]

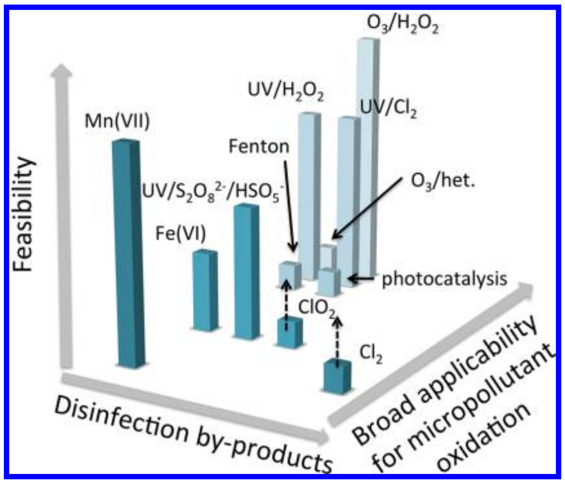


Figure 1. Comparison of various chemical oxidants/processes for the abatement of micropollutants in municipal water treatment. Feasible processes are in the upper left corner (low DBP formation, oxidation of a broad range of micropollutants and a high feasibility (maturity and low energy demand). The arrows for chlorine dioxide and chlorine show that their feasibility is higher than shown in the picture, because they are widely applied in water treatment. Example how to read the graph: Mn(VII) has low formation of DBPs, a high feasibility (already widely applied in water treatment) but is a very selective oxidant, which does not warrant oxidation of a wide range of micropollutants.

Transformation Products and Effects. The number of studies on the formation of oxidative transformation products increased significantly in the early 2000s concomitantly with the development of analytical methods such as liquid chromatography (LC) coupled with high resolution mass spectrometry.[5,83] This resulted in a significant mechanistic knowledge gain for oxidation processes involving $^{\bullet}OH$[84,85] and O_3,[27,86,87] which enabled the compilation of pathway prediction systems for transformation reactions induced by $^{\bullet}OH$[88] and O_3.[89] Researchers also studied ferrate,[51,90,91] chlorine,[26,92−106] bromine,[107,108] permanganate[109] and chlorine dioxide,[110−114] adding to the mechanistic understanding. This wealth of information triggered new worries about the formation of potentially toxic transformation products.[115] Attempts to couple mechanistic information to toxicological consequences led to three major cases for transformation products that can be distinguished (Figure 2):

Oxidative Treatment Leads to a Loss of the Primary Biological Activity Caused by the Parent Compound (Lower Red Curve in Figure 2). Numerous toxicological studies have demonstrated that the primary or secondary attack of an oxidant (most studies for O_3 and/or $^{\bullet}OH$) on a target compound leads to a proportional loss of the primary biological effects for various classes of compounds such as antimicrobial agents,[116−121] estrogenic compounds,[122−124] herbicides[125] and insecticides.[126]

The Transformation Products of Biologically Active Molecules Are (Potentially) Regulated Compounds with Higher Toxicity than the Parent Compound (Upper Red Curve in Figure 2). A case in point is the formation of chloroform during chlorination of triclosan, an antimicrobial agent.[105,127] Even though in this case the antimicrobial activity of the target compound is lost, a new effect arises. This problem is even aggravated, by the photochemical formation of dioxins from chlorinated triclosan released to the aquatic environment.[106] Another example is the formation of NDMA during chloramination of ranitidine-containing water.[128,129]

Formation of Toxic/Bioactive Compounds from Target Compounds with Low Biological Activity (Upper Red Curve in Figure 2). *N*,*N*-dimethylsulfamide, a nontoxic metabolite of the fungicide tolylfluanide, is transformed to the genotoxic *N*-nitrosodimethylamine (NDMA) during ozonation.[130,131] NDMA was also formed during ozonation of secondary wastewater effluents containing antiyellowing agents.[132] In another example the attack of $^{\bullet}OH$ on quinoline led to an estrogenic compound, however, with a very low potency.[133]

THE OXIDATION DILEMMA

Oxidant Reactions with DOM. For simplicity, most mechanistic studies on contaminant oxidation are conducted in ultrapure water. However, under realistic conditions, the largest fraction of chemical oxidants is consumed by DOM (Figure 3).[134−136] Much of this is due to oxidation of phenolic

DOI: 10.1021/acs.est.8b00586
Environ. Sci. Technol. 2018, 52, 5062−5075

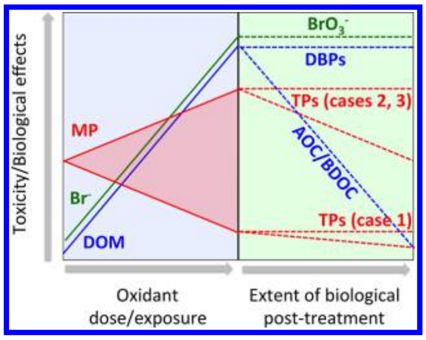


Figure 2. Relative evolution of toxicity (human health) and/or biological effects during oxidative treatment of micropollutant-containing waters as a function of the oxidant dose/exposure and the extent of biological post-treatment. MP: micropollutant; DOM: dissolved organic matter; TPs: Transformation products; DBPs: Disinfection byproducts; AOC: Assimilable organic carbon; BDOC: Biodegradable organic carbon.

moieties in DOM (1−2 mmol/gDOC, that is, μM DOM-phenol concentrations).[137] This is much higher than micropollutant concentrations of ≪0.1 μM in natural waters and wastewater effluents.[30,138]

The reaction of DOM with ozone and/or $^\bullet$OH leads to the formation of oxygen-rich byproducts (e.g., aldehydes, ketones and carboxylic acids) in total concentrations often >100 μg/L,[139,140] which is significantly above the sum of the concentrations of micropollutants (ca. 25 μg/L) detected in a wastewater treatment plant effluent after ozonation.[138] Therefore, oxygen-rich byproducts may contribute more to the toxicity/biological effects of the treated water than micropollutants (Figure 2, blue line).[141] Cases in point are the formation of toxic α,β-unsaturated enedials and oxoenals during UV treatment of phenols in presence or absence of H_2O_2 or the formation of quinones or catechols during ozonation.[142−144] In line with the formation of such products, it has been observed that the toxicity to certain aquatic organisms increases after wastewater ozonation, with mostly an improvement after a biological treatment step (Figure 2, dashed blue line).[141,145−147] Since these oxygen-rich byproducts are often easily biodegradable, a biological post-treatment step is routinely implemented after an oxidation/AOP in drinking water and wastewater effluents.[11,138−141,148−150] So far, there is not sufficient information to assess the efficiency of biological post-treatment for transformation product abatement. However, it has been shown that various nitrogenous transformation products (e.g., *N*-oxides) are not removed in biological post-treatment after ozonation of a municipal wastewater effluent (Figure 2, horizontal dashed red lines).[138] Nevertheless, it can be expected that ozone transformation products of aromatic compounds or olefins are more biodegradable than their parent compounds (Figure 2, descending dashed red lines).[86]

Bromide in Oxidative Water Treatment. Bromide plays a special role in oxidative water treatment because it can be oxidized to bromine, a reactive secondary oxidant, by some common oxidants.[107] Bromine reacts with DOM to produce brominated byproducts (Br-DBPs), which are typically more toxic than the chlorinated analogues.[151,152] If these Br-DBPs are formed, they are mostly recalcitrant during biological post-treatment (Figure 2, horizontal dashed blue line). For advanced oxidation processes such as O_3/H_2O_2 or UV/H_2O_2, these DBPs are less important, because the transient bromine concentration is typically low and the bromine-reactive precursors are consumed by ozone and/or $^\bullet$OH. The major drawback for ozone-based treatment of bromide-containing waters is the formation of bromate,[153] which is a possible human carcinogen with drinking water and ecotoxicological standards of 10 and 50 μg/L bromate, respectively.[154,155] Bromate removal (e.g., by biological post-treatment) after its formation is not a viable option (Figure 2, dashed green line) and its formation has to be minimized/avoided as much as possible.[9,27,153,154,156−164] Compared to conventional ozonation, the O_3/H_2O_2 process leads to a significantly lower bromate formation.[157]

Trade-off between Energy Consumption and Bromate. In contrast to ozone-based processes, the UV/H_2O_2 process does not lead to bromate formation in bromide-containing waters (Supporting Information (SI) Figure S1).[157,165] However, the energy consumption for the abatement of micropollutants to 10% of their initial concentrations, the electrical energy per order (EEO), is generally about 4−20 times higher (0.17−2.3 kWh/m^3) for the UV/H_2O_2 process than for the O_3/H_2O_2 process (0.04−0.25 kWh/m^3).[166,167] Therefore, there is a trade-off between bromate formation and energy consumption, O_3/H_2O_2 is favored at lower and UV/H_2O_2 at higher bromide concentrations.[168] If disinfection is an additional criteria, the UV/H_2O_2 process has a superior performance, because the high UV fluences for this AOP lead to an efficient inactivation of microorganisms (SI Figure S1).[167] In contrast, only limited inactivation can be achieved by the O_3/H_2O_2 process, because the ozone exposure is reduced due to its hydrogen peroxide induced decomposition to $^\bullet$OH, which is a weak disinfectant.[27,169]

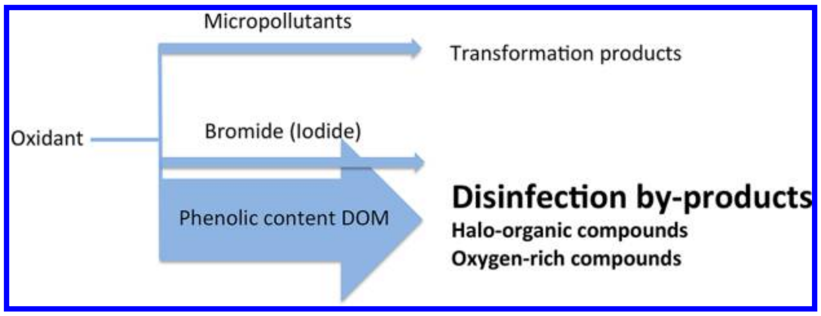


Figure 3. Relative contribution of the various water solutes to the consumption of oxidants. The DOM is by far the major consumer of oxidants during water treatment, followed by bromide (and in some cases iodide) depending on their concentrations. Compared to these matrix components, micropollutants only contribute minimally to the oxidant consumption. Disinfection byproducts are formed from the reactions of oxidants with the matrix components, whereas the oxidation of micropollutants leads to transformation products.

AN UNEXPECTED REVIVAL OF OXIDATION PROCESSES WITH A SIGNIFICANT KNOWLEDGE GAIN

Enhanced Wastewater Treatment. After a research and development phase of almost two decades,[170−172] the Swiss authorities decided to upgrade about 15% of the country's municipal wastewater treatment plants to reduce the load of organic micropollutants to the aquatic environment.[173−177] This development was triggered by detection of pharmaceuticals, endocrine disruptors and personal care products in municipal wastewater effluents,[178−182] combined with ecological studies suspecting/demonstrating the effect of endocrine disruptors on fish populations.[183−187] In Germany and France similar approaches are currently being assessed.[145,147,188−190] Overall, the European approach is guided by mostly ecological concerns, with an added benefit for drinking water, which is often abstracted from rivers (i.e., riverbank filtration) downstream of wastewater effluent discharges.

Water Reuse. The recent increase in municipal water reuse is mainly driven by water security (e.g., Singapore[191]) and water scarcity in arid areas, which is often aggravated by climate change, population growth and competition for water resources with agriculture.[192,193] A case in point is California, where water was initially recycled for industrial and agricultural purposes and then for drinking water production.[194,195] Recently, a rigorous framework of multibarrier advanced water treatment systems was implemented to fulfill the criteria for direct water reuse and the surface water treatment rule.[194,196−198] All of the suggested direct potable reuse treatment trains in California include an ozonation and/or UV/H_2O_2 process combined with pre- or post-treatment with reverse osmosis for disinfection and the abatement of micropollutants.[194]

Decentralized Systems. In the future, centralized municipal water systems may be partially replaced by more localized systems, due to the high cost of maintaining aging infrastructure of drinking water and wastewater networks as well as the rapid development of megacities. To support low maintenance water treatment in decentralized systems, electricity-based systems will likely prove to be advantageous both for disinfection and for oxidation of micropollutants.[193,199−201]

Finding the Needle in the Haystack. *The Analytical Approach.* The uncertainty about transformation products being formed during oxidation might become a barrier to these developments as already shown for the atrazine example above. The application of LC-high resolution MS-based analytical methods and data processing tools led to the detection of hundreds of micropollutants and their biological/oxidative transformation products in wastewater matrices.[138,202−204] Currently, the interpretation of such data sets is very limited and we need a better connection to mechanistic studies and toxicity databases. The detection of new compounds often triggers kinetic and mechanistic studies of oxidation reactions of randomly selected micropollutants, which only leads to limited progress in the field. Rather, the emphasis should be on fundamental studies on selected functional groups, which are transferrable to micropollutants containing the same chemical moieties.[27] Several promising approaches are discussed below.

Prediction of Reaction Kinetics for Oxidation Reactions. The efficiency of an oxidative treatment depends directly on the reaction kinetics. There is already a large kinetic database for the reactions of various oxidants such as $^{\bullet}OH$,[205] ozone,[27] chlorine,[26] chlorine dioxide,[206] bromine[107] with organic micropollutants. In addition, several approaches, such as quantitative structure activity relationships (QSARs), quantum chemical computations (reactions with ozone or $^{\bullet}OH$) and the group contribution method ($^{\bullet}OH$) have been applied to predict second order rate constants for the reactions of organic compounds with oxidants.[26,63,107,207−211] These empirical correlations yield predictions within a factor of 3 of the measured values,[208] which is within the range of variation in measured second order rate constants from different laboratories.[27]

Prediction of Transformation Products. In addition to understanding the kinetics of compound abatement, the elucidation of transformation pathways for oxidative treatment of micropollutants is a tedious process involving labor-intense experiments with product analysis by various methods (e.g., GC-MS/MS, LC-MS/MS, NMR, etc.) (Figure 4a).[27,76−78,85,86,108,110,114,117,212−224] Identification of the products of 10% of the 100 000 most common products in commerce[145] could cost over 100 million USD for each oxidant assuming a very optimistic average time of 3 months per compound for a product study. This cost would be augmented by the thousands of new chemicals, which are registered every year.[145] This theoretical exercise demonstrates that such a comprehensive screening is unlikely to occur. Instead, we need a prioritization and fundamental studies with model com-

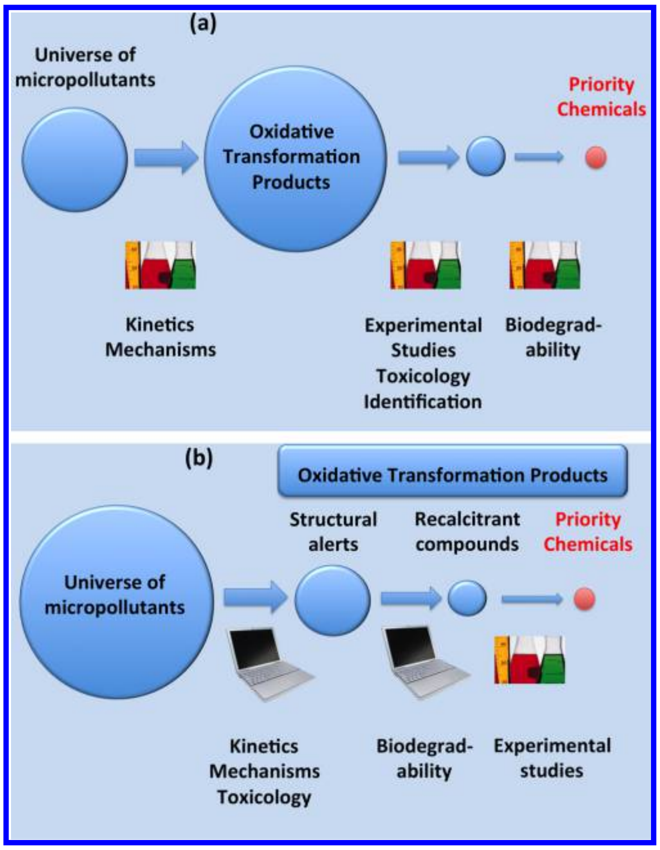


Figure 4. Two approaches dealing with large numbers of compounds during oxidation processes. (a) Experimental approach with kinetic and mechanistic studies, toxicological assessment and elucidation of problematic compounds by effect-driven analyses. (b) In silico assessment of micropollutants by prediction of transformation products and potential (eco)toxicological risks. For (b), experimental studies will only be performed with critical compounds.

pounds, which can be extrapolated to the vast number of detected compounds. To compile and explore such data in a comprehensive manner, tiered screening methods are needed (Figure 4b). Such approaches have been used for biological transformation products of organic chemicals over the last decades.[225−228] Similarly, pathway prediction platforms have been developed recently for $^{\bullet}OH$[88] and for ozone reactions.[89] The latter platform determines second order rate constants for the reactions of ozone-reactive moieties and predicts potential products based on >100 reaction rules.[89] This allows an interpretation of results from nontarget analyses of oxidation processes[229] and a rapid screening of possible transformation products for potentially toxic structures for, e.g., known carcinogenic groups (Figure 4b).[230,231] There are numerous in silico prediction methods for mutagenicity, modes of action and ecotoxicity, which have been implemented in software packages.[232−234] Only after a predicted compound is identified and suspected to cause a potential risk, it will be necessary to conduct laboratory studies on its formation, stability, fate and toxicity.

■ SEARCHING FOR THE SILVER BULLET

Future urban water systems may include both centralized and decentralized components with water reuse of e.g., gray water and concentrate streams.[193] Whereas, centralized water systems will probably rely on well-established oxidation/disinfection methods, in decentralized systems (point-of-entry/use) electricity-based methods such as UV irradiation and electrochemical production of oxidants such as chlorine, hydrogen peroxide and ozone bear a huge potential because they are independent of the transport of chemicals.[201,235−237] In recent years, the portfolio of chemical oxidation has been expanded to search for higher energy efficiency and lower DBP formation. Often, combined/hybrid processes are tested, however, an imperative comparison to the current benchmark oxidation processes ozonation, O_3/H_2O_2 and UV/H_2O_2 is often missing.

Ozone-Based Processes. Heterogeneous ozonation processes have been widely studied to enhance the transformation of ozone to hydroxyl radicals.[238] A recently developed catalyst was efficient in reducing bromate formation with an efficient abatement of ozone-resistant micropollutants such as atrazine.[239] However, the optimal concentration of the catalyst of 0.5 g/L (500g/m^3) causes substantial problems related to the process design. The potential gain in efficiency is impaired by the problems related to heterogeneous processes, such as separation, fouling and deactivation of the catalyst, bleeding of potentially toxic elements and costs of the catalysts. Furthermore, natural and H_2O_2-induced ozone decomposition have hydroxyl radical yields of about 50%,[27,240] wherefore, the margin for improvement is only about a factor of 2 if a process leads to 100% transformation of ozone to hydroxyl radical.

Hence, the homogeneous O_3/H_2O_2 process seems a more reasonable approach for bromate control by keeping the dissolved ozone concentration very low.[241]

To this end, optimized processes involving membranes (SI Figure S2a), multiple static mixers or multiple injection ports for ozone transfer to hydrogen peroxide-containing waters can be applied to warrant very low transient ozone concentrations leading to almost neglect bromate formation.[241−243] Hydrogen peroxide can be produced electrochemically onsite and on demand as in the electro-peroxone process (SI Figure S2a),[244] avoiding the transport of chemicals and enhancing the flexibility of the process.

UV-Based Processes. Vacuum UV (VUV, UV radiation <200 nm) has been studied as a chemical free AOP for decades.[245] However, its application at scale has been limited because only a thin layer of water can be treated with an EEO of about 1 kWh/m^3 for micropollutant abatement.[246] An interesting application with a reduced energy demand could be a combination of VUV irradiation with ozone formed in the gas phase (SI Figure S2b)[246] Alternatively, a combined VUV and UV system yielding a relatively energy efficient abatement of atrazine, similar to a conventional UV/H_2O_2 process was used for treatment of a few L/min,[167,247] making it interesting for a combined disinfection/oxidation in point-of-use systems.

Recently, the UV/chlorine process has gained a lot of attention. In this process, the photolysis of chlorine yields hydroxyl radicals, chlorine atoms and ozone, which may lead to a more efficient abatement of micropollutants and inactivation of chlorine-resistant microorganisms.[248−252] The formation of organic chlorinated DBPs does not seem to be a major issue, but inorganic DBPs such as chlorate and in bromide-containing waters, bromate, are formed at significant levels and have to be carefully evaluated before implementing this process.[248,253,254] The UV/chlorine process is more efficient for micropollutant abatement compared to UV/H_2O_2 at low pH (i.e., 5.5) in reverse osmosis permeates in water reuse systems, whereas it is less efficient in the near neutral pH range.[255] The UV/chloramine process seems to be an interesting variant in the context of water reuse, where chloramines are already present to avoid RO membrane fouling.[255,256]

LED-Based Processes. Some UV-based processes such as visible-to-UVC up-conversion and photovoltaics-driven or standalone UV-light emitting diodes (LED) are in development. Among different approaches numerous investigators have speculated about the advantages of UV/LEDs for disinfection in small-scale point-of-use/entry systems.[257−260] Even though the development of UV-LEDs has made significant progress in the past decade, the energy efficiency is still quite low (i.e., in the order of 1 to a few percent).[261−263] The further development of these devices will almost certainly be driven by applications other than water treatment due to the relatively small size of the current market.[264] Furthermore, the lifetime of UV LED, a major advantage of visible LEDs, is still very limited. For visible LEDs, a price drop of a factor of 10 per decade with a 20-fold increase of photon flux per lamp occurred. This development jump was possible due to the application of LEDs as LCD backlights.[265] Overall, it is unlikely, that UV/LEDs will play a major role in water treatment over the next two decades without a substantial research and development effort.

Electrochemical Processes. Electrochemical processes have undergone a revival in recent years, due to the promise for transformation of chemicals just by electricity.[266] However, there are still many obstacles to overcome, such as the formation of oxidation byproducts, undesired side reactions, mass transfer limitations and high energy demand, which may prevent their application in large-scale water treatment.[235,266] However, there seems to be potential for small-scale onsite treatment applications. One example is a combined electrochemical-UV system, which uses hydrogen peroxide produced in a gas diffusion cathodic cell in a UV/H_2O_2 reactor and the residual hydrogen peroxide is destroyed by chlorine, produced at the anode after the AOP (SI Figure S2c).[201] Although, this system needs some optimization, it illustrates the great potential of electrochemical processes to achieve oxidation, disinfection, and to provide a residual disinfectant after treatment.

In contrast, the oxidation of micropollutants by hydroxyl radicals and other reactive species formed on electrode surfaces, such as boron-doped diamond (BDD) electrodes, has low efficiency due to the diffusion limitations, competition with DOM and electrode surface limitation.[267] In addition, costs and electrode stability are still an issue for BDDs.

Plasma, Sonolysis, Natural and Synthetic Enzymes. Plasma-based and sonolysis processes have been studied for water treatment for several decades. Although they can oxidize contaminants,[268] cost-effective applications seem to be far away or even impossible due to their high energy demand and significant up-scaling issues.[33,269–272] Recently, ozone generators based on microplasma have become available and seem to be an attractive alternative at small-scale to conventional ozone generators.[273]

The application of natural and synthetic enzymes has been suggested as a sustainable option for water treatment.[274,275] However, there are still limitations to these processes (see SI Text 1).

■ OXIDATION PROCESSES: THE WAY FORWARD

The dilemma of water treatment processes not being selective to specific target compounds is a severe limitation and a practical inevitability for oxidation processes. The ubiquitous matrix components reduce the treatment efficiency during oxidation and may lead to unintended consequences, such as disinfection byproducts. Furthermore, micropollutants are not removed during oxidation but transformed to a multitude of products. During sequential transformation processes, the concentration of individual transformation products decreases (oxidative dilution) but in some cases the transformation products are more toxic than the parent compounds.

How do we guarantee that the risks of this approach do not outweigh the benefits? Unfortunately, the risk cannot be entirely eliminated, however, it can be minimized by a rigorous assessment approach. The following questions need to be addressed for an optimum application of oxidation processes.

Will the Oxidation Process Achieve the Needed Decrease in Micropollutant Concentrations? Kinetics is key to the efficiency of an oxidation process. With existing knowledge, we can estimate oxidation kinetics, which is mostly sufficient to estimate the oxidative abatement efficiency in real systems. For novel oxidation processes, the overall energy demand and operating costs need to be similar or below benchmarked processes such as chlorination, ozonation or UV-based processes. The efficiency of novel processes tends to improve as technologies are further developed. For distributed treatment systems, a higher energy demand is justifiable, because no/minimal energy-intensive transport of water is required in these cases.

Research needs: Further development of kinetics prediction tools based on quantum chemical computations for existing and emerging oxidation technologies.

What Is the Risk of the Formation of Toxic Transformation Products? For certain oxidants and functional groups, transformation product formation can be assessed by computer-based prediction tools. A combination of these tools with advanced analytical methods allows the elucidation of transformation products and their potential toxicological consequences.

Research needs: The focus of further experimental research should be based on missing basic information related to functional groups and oxidants, which is applicable to whole classes of micropollutants. Improved workflows and data analysis for target and nontarget analyses are required and we need a better understanding of what is formed from the oxidation of micropollutants versus the organic matrix. Sensitive, robust, standardized and high throughput toxicological test systems with various human toxicological and ecotoxicological end points need to be developed.

What Are the Implications of Oxidation of the Matrix Components? Due to the variability and complexity of natural or effluent DOM, the prediction of DBPs is still in its infancy.

Research needs: Quantification of reactive functional groups in DOM for a better prediction of the efficiency of oxidation and of the formation of ensuing products.

Which Is the Best Oxidative Treatment Option? Based on the complexity of oxidation processes, there is a need for broadband and tailored oxidation processes combined with other treatments, to remove transformation products and take advantage of multiple barriers. Mostly biological and/or adsorptive post-treatment after oxidation should be applied, because of their benefits for abatement of oxygenated DBPs and/or transformation products. The portfolio of oxidation processes should consider applications at various scales from municipal systems to single users and recognize the way that other water quality aspects (e.g., bromide) affect the performance.

Research needs: Further development of electricity-driven processes, which avoid transport and storage of chemicals and are applicable at various scales. Furthermore, there is a need for improved engineering design to allow scaling up of these processes. Development of better models for activated carbon adsorption and further developments of computer-based prediction tools to assess biological transformation of disinfection byproducts and transformation products.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b00586.

Additional information as noted in the text(PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: urs.vongunten@eawag.ch.

ORCID

Urs von Gunten: 0000-0001-6852-8977

Notes

The author declares no competing financial interest.

Biography

Photo provided by Urs von Gunten

Urs von Gunten is an environmental chemist, who has more than 25 years of experience in oxidative water and wastewater treatment processes. His main interests are in kinetic and mechanistic studies of oxidatively-induced transformation processes of micropollutants and the formation of disinfection by-products from matrix components and the ensuing (eco)toxicological consequences. Besides his academic activities, he emphasizes collaborations with practitioners from the water sector to transfer the findings of his research to real-world applications.

■ ACKNOWLEDGMENTS

I thank David Sedlak for many fruitful discussions and inspiring inputs during my sabbatical at UC Berkeley.

■ REFERENCES

(1) McGuire, M. J. Eight revolutions in the history of US drinking water disinfection. *J. - Am. Water Works Assoc.* **2006**, *98* (3), 123−149.

(2) Le Paulouë, J.; Langlais, B. State of the art of ozonation in France. *Ozone: Sci. Eng.* **1999**, *21*, 153−162.

(3) Bellar, T. A.; Lichtenberg, J. J.; Kroner, R. C. The Occurrence of Organohalides in Chlorinated Drinking Waters. *J. - Am. Water Works Assoc.* **1974**, *66* (12), 703−706.

(4) Rook, J. J. Formation of haloforms during chlorination of natural waters. *Water Treat. Exam.* **1974**, *23*, 234−243.

(5) Richardson, S. D.; Kimura, S. Y. Water Analysis: Emerging Contaminants and Current Issues. *Anal. Chem.* **2016**, *88* (1), 546−582.

(6) Sedlak, D. L.; von Gunten, U. The Chlorine Dilemma. *Science* **2011**, *331* (6013), 42−43.

(7) Hrudey, S. E. Chlorination disinfection by-products, public health risk tradeoffs and me. *Water Res.* **2009**, *43* (8), 2057−2092.

(8) Li, X.-F.; Mitch, W. A. Drinking Water Disinfection Byproducts (DBPs) and Human Health Effects: Multidisciplinary Challenges and Opportunities. *Environ. Sci. Technol.* **2018**, *52* (4), 1681−1689.

(9) von Gunten, U. Ozonation of drinking water: Part II. Disinfection and by-product formation. *Water Res.* **2003**, *37*, 1469−1487.

(10) Hammes, F.; Meylan, S.; Salhi, E.; Koster, O.; Egli, T.; Von Gunten, U. Formation of assimilable organic carbon (AOC) and specific natural organic matter (NOM) fractions during ozonation of phytoplankton. *Water Res.* **2007**, *41* (7), 1447−1454.

(11) Van der Kooij, D.; Hijnen, W. A. M.; Kruithof, J. The effects of ozonation, biological filtration and the distribution on the concentration of easily assimilable organic carbon (AOC) in drinking water. *Ozone: Sci. Eng.* **1989**, *11*, 297.

(12) Ramseier, M. K.; Peter, A.; Traber, J.; von Gunten, U. Formation of assimilable organic carbon during oxidation of natural waters with ozone, chlorine dioxide, chlorine, permanganate, and ferrate. *Water Res.* **2011**, *45* (5), 2002−2010.

(13) Aieta, E. M.; Berg, J. D. A REVIEW OF CHLORINE DIOXIDE IN DRINKING-WATER TREATMENT. *J. - Am. Water Works Assoc.* **1986**, *78* (6), 62−72.

(14) Krasner, S. W.; Mitch, W. A.; McCurry, D. L.; Hanigan, D.; Westerhoff, P. Formation, precursors, control, and occurrence of nitrosamines in drinking water: A review. *Water Res.* **2013**, *47* (13), 4433−4450.

(15) Mitch, W. A.; Sharp, J. O.; Trussell, R. R.; Valentine, R. L.; Alvarez-Cohen, L.; Sedlak, D. L. N-nitrosodimethylamine (NDMA) as a drinking water contaminant: A review. *Environ. Eng. Sci.* **2003**, *20* (5), 389−404.

(16) Gordon, G.; Slootmaekers, B.; Tachiyashiki, S.; Wood, D. W. Minimizing Chlorite Ion and Chlorate Ion in Water Treated with Chlorine Dioxide. *J. - Am. Water Works Assoc.* **1990**, *82* (4), 160−165.

(17) Richardson, S. D.; Plewa, M. J.; Wagner, E. D.; Schoeny, R.; DeMarini, D. M. Occurrence, genotoxicity, and carcinogenicity of regulated and emerging disinfection by-products in drinking water: A review and roadmap for research. *Mutat. Res., Rev. Mutat. Res.* **2007**, *636* (1−3), 178−242.

(18) WHO *Guidelines for Drinking Water Quality*; World Health Organization: Geneva, 2017.

(19) Bruchet, A.; Duguet, J. P. Role of oxidants and disinfectants on the removal, masking and generation of tastes and odours. *Rev. Environ. Sci. Bio/Technol.* **2004**, *49* (9), 297−306.

(20) Piriou, P.; Soulet, C.; Acero, J. L.; Bruchet, A.; Von Gunten, U.; Suffet, I. H. Understanding medicinal taste and odour formation in drinking waters. *Water Sci. Technol.* **2007**, *55* (5), 85−94.

(21) Rosario-Ortiz, F.; Rose, J.; Speight, V.; von Gunten, U.; Schnoor, J. How do you like your tap water? *Science* **2016**, *351* (6276), 912−914.

(22) Yu, W.; Campos, L.; Shi, T.; Li, G.; Graham, N. Enhanced removal of manganese in organic-rich surface water by combined sodium hypochlorite and potassium permanganate during drinking water treatment. *RSC Adv.* **2015**, *5* (35), 27970−27977.

(23) Allard, S.; Fouche, L.; Dick, J.; Heitz, A.; von Gunten, U. Oxidation of Manganese(II) during Chlorination: Role of Bromide. *Environ. Sci. Technol.* **2013**, *47* (15), 8716−8723.

(24) Sain, A. E.; Dietrich, A. M. Rethinking aesthetic guidelines for manganese and iron in drinking water. *Aqua* **2015**, *64* (7), 775.

(25) Rice, R. G. Ozone in the United States of America - state of the art. *Ozone: Sci. Eng.* **1999**, *21*, 99−118.

(26) Deborde, M.; von Gunten, U. Reactions of chlorine with inorganic and organic compounds during water treatment - Kinetics and mechanisms: A critical review. *Water Res.* **2008**, *42* (1−2), 13−51.

(27) von Sonntag, C.; von Gunten, U. *Chemistry of Ozone in Water and Wastewater Treatment. From Basic Principles to Applications*; IWA: London, 2012.

(28) Wolfe, R. L.; Ward, J. F.; Olson, G. R. Inorganic Chloramines as Drinking Water Disinfectants: A Review. *J. - Am. Water Works Assoc.* **1984**, *77* (5), 74−88.

(29) Schwarzenbach, R. P.; Egli, T.; Hofstetter, T. B.; von Gunten, U.; Wehrli, B. Global Water Pollution and Human Health. *Annual Review of Environment and Resources* **2010**, *35*, 109−136.

(30) Schwarzenbach, R. P.; Escher, B. I.; Fenner, K.; Hofstetter, T. B.; Johnson, C. A.; von Gunten, U.; Wehrli, B. The challenge of micropollutants in aquatic systems. *Science* **2006**, *313* (5790), 1072−1077.

(31) von Gunten, U. Ozonation of drinking water: Part I. Oxidation kinetics and product formation. *Water Res.* **2003**, *37*, 1443−1467.

(32) Kruithof, J. C.; Kamp, P. C.; Martijn, B. J. UV/H2O2 treatment: A practical solution for organic contaminant control and primary disinfection. *Ozone: Sci. Eng.* **2007**, *29* (4), 273−280.

(33) von Sonntag, C. Advanced oxidation processes: mechanistic aspects. *Water Sci. Technol.* **2008**, *58* (5), 1015−1021.

(34) Lee, M.; Merle, T.; Rentsch, D.; Canonica, S.; von Gunten, U. Abatement of Polychoro-1,3-butadienes in Aqueous Solution by Ozone, UV Photolysis, and Advanced Oxidation Processes (O3/H2O2 and UV/H2O2). *Environ. Sci. Technol.* **2017**, *51* (1), 497−505.

(35) Guan, X. H.; He, D.; Ma, J.; Chen, G. H. Application of permanganate in the oxidation of micropollutants: a mini review. *Front. Environ. Sci. Eng. China* **2010**, *4* (4), 405−413.

(36) Waldemer, R. H.; Tratnyek, P. G. Kinetics of Contaminant Degradation by Permanganate. *Environ. Sci. Technol.* **2006**, *40* (3), 1055−1061.

(37) Sun, B.; Dong, H.; He, D.; Rao, D.; Guan, X. Modeling the Kinetics of Contaminants Oxidation and the Generation of Manganese(III) in the Permanganate/Bisulfite Process. *Environ. Sci. Technol.* **2016**, *50* (3), 1473−1482.

(38) Sun, B.; Guan, X.; Fang, J.; Tratnyek, P. G. Activation of Manganese Oxidants with Bisulfite for Enhanced Oxidation of Organic Contaminants: The Involvement of Mn(III). *Environ. Sci. Technol.* **2015**, *49* (20), 12414−12421.

(39) Miller, C. J.; Wadley, S.; Waite, T. D., Fenton, photo-Fenton and Fenton-like processes. In *Advanced Oxidation for Water Treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publisher: London, 2018.

(40) He, X.; de la Cruz, A. A.; O'Shea, K. E.; Dionysiou, D. D. Kinetics and mechanisms of cylindrospermopsin destruction by sulfate radical-based advanced oxidation processes. *Water Res.* **2014**, *63*, 168−178.

(41) Lutze, H. V.; Bircher, S.; Rapp, I.; Kerlin, N.; Bakkour, R.; Geisler, M.; von Sonntag, C.; Schmidt, T. C. Degradation of Chlorotriazine Pesticides by Sulfate Radicals and the Influence of Organic Matter. *Environ. Sci. Technol.* **2014**, *49* (3), 1673−1680.

(42) Ghanbari, F.; Moradi, M. Application of peroxymonosulfate and its activation methods for degradation of environmental organic pollutants: Review. *Chem. Eng. J.* **2017**, *310* (Part 1), 41−62.

(43) Ye, T.; Wei, Z.; Spinney, R.; Tang, C.-J.; Luo, S.; Xiao, R.; Dionysiou, D. D. Chemical structure-based predictive model for the oxidation of trace organic contaminants by sulfate radical. *Water Res.* **2017**, *116*, 106−115.

(44) Wacławek, S.; Lutze, H. V.; Grübel, K.; Padil, V. V. T.; Černík, M.; Dionysiou, D. D. Chemistry of persulfates in water and wastewater treatment: A review. *Chem. Eng. J.* **2017**, *330* (Supplement C), 44−62.

(45) Luo, S.; Wei, Z.; Dionysiou, D. D.; Spinney, R.; Hu, W.-P.; Chai, L.; Yang, Z.; Ye, T.; Xiao, R. Mechanistic insight into reactivity of sulfate radical with aromatic contaminants through single-electron transfer pathway. *Chem. Eng. J.* **2017**, *327* (SupplementC), 1056−1065.

(46) Zhang, B.-T.; Zhang, Y.; Teng, Y.; Fan, M. Sulfate Radical and Its Application in Decontamination Technologies. *Crit. Rev. Environ. Sci. Technol.* **2015**, *45* (16), 1756−1800.

(47) Lutze, H. V.; Brekenfeld, J.; Naumov, S.; von Sonntag, C.; Schmidt, T. C. Degradation of perfluorinated compounds by sulfate radicals − New mechanistic aspects and economical considerations. *Water Res.* **2018**, *129*, 509−519.

(48) Tsitonaki, A.; Petri, B.; Crimi, M.; MosbÆK, H.; Siegrist, R. L.; Bjerg, P. L. In Situ Chemical Oxidation of Contaminated Soil and Groundwater Using Persulfate: A Review. *Crit. Rev. Environ. Sci. Technol.* **2010**, *40* (1), 55−91.

(49) Siegrist, L. R.; Crimi, M.; Simpkin, T. J. *In Situ Chemical Oxidation for Groundwater Remediation*; Springer: New York, 2011.

(50) Jiang, J.-Q.; Durai, H. B. P.; Winzenbacher, R.; Petri, M.; Seitz, W. Drinking water treatment by in situ generated ferrate(VI). *Desalin. Water Treat.* **2015**, *55* (3), 731−739.

(51) Sharma, V. K.; Zboril, R.; Varma, R. S. Ferrates: Greener Oxidants with Multimodal Action in Water Treatment Technologies. *Acc. Chem. Res.* **2015**, *48* (2), 182−191.

(52) Reddy, P. A. K.; Reddy, P. V. L; Kwon, E.; Kim, K.-H.; Akter, T.; Kalagara, S. Recent advances in photocatalytic treatment of pollutants in aqueous media. *Environ. Int.* **2016**, *91* (91), 94−103.

(53) Lee, S.-Y.; Park, S.-J. TiO2 photocatalyst for water treatment applications. *J. Ind. Eng. Chem.* **2013**, *19* (6), 1761−1769.

(54) Chong, M. N.; Jin, B.; Chow, C. W. K.; Saint, C. Recent developments in photocatalytic water treatment technology: A review. *Water Res.* **2010**, *44* (10), 2997−3027.

(55) Dong, H.; Zeng, G.; Tang, L.; Fan, C.; Zhang, C.; He, X.; He, Y. An overview on limitations of TiO2-based particles for photocatalytic degradation of organic pollutants and the corresponding countermeasures. *Water Res.* **2015**, *79*, 128−146.

(56) Wilson, E. TiO2 appears inefficient for water treatment. *Chem. Eng. News* **1996**, *74* (27), 29.

(57) Cates, E. L. Photocatalytic Water Treatment: So Where Are We Going with This? *Environ. Sci. Technol.* **2017**, *51* (2), 757−758.

(58) Stancl, H. O. N.; Hristovski, K.; Westerhoff, P. Hexavalent Chromium Removal Using UV-TiO2/Ceramic Membrane Reactor. *Environ. Eng. Sci.* **2015**, *32* (8), 676−683.

(59) Pillali, S. C.; McGuiness, N. B.; Byrne, C.; Han, C.; Lalley, J.; Nadagouda, M.; Falares, P.; Kontos, A. G.; Garcia-Pinilla, M. A.; O'Shea, K.; Mangalaraja, R. V.; Christophoridis, C.; Triantis, T.; Hiskia, A.; Dionysiou, D. D., Photocatalysis as an effective advanced oxidation process. In *Advanced Oxidation Processes for Water treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publisher: London, 2018.

(60) Lee, Y.; von Gunten, U. Oxidative transformation of micropollutants during municipal wastewater treatment: Comparison of kinetic aspects of selective (chlorine, chlorine dioxide, ferrateVI, and ozone) and non-selective oxidants (hydroxyl radical). *Water Res.* **2010**, *44* (2), 555−566.

(61) Loeb, B. L.; Thompson, C. M.; Drago, J.; Takahara, H.; Baig, S. In *Worldwide Ozone Capacity for Treatment of Drinking Water and Wastewater: A Review*; IOA IUVA World Congress & Exhibition:Paris, b 2011; pp VII.2−1−VII.2 −20.

(62) Geering, F. Ozone application: the state of the art in Switzerland. *Ozone: Sci. Eng.* **1999**, *21*, 187−200.

(63) Hoigné, J.; Bader, H. Rate constants of reactions of ozone with organic and inorganic compounds in water. I. Non-dissociating organic compounds. *Water Res.* **1983**, *17*, 173−183.

(64) Hoigne, J.; Bader, H. Rate Constants of Reactions of Ozone with Organic and Inorganic-Compounds in Water 2. Dissociating Organic-Compounds. *Water Res.* **1983**, *17* (2), 185−194.

(65) Pryor, W. A.; Giamalva, D. H.; Church, D. F. Kinetics of Ozonation. 2. Amino Acids and Model Compounds in Water and Comparisons to Rates in Nonpolar Solvents. *J. Am. Chem. Soc.* **1984**, *106*, 7094−7100.

(66) Lee, G. F.; Morris, J. C. Kinetics of chlorination of phenol - chlorophenolic tastes and odors. *Int. J. Air Waer. Pollut.* **1962**, *6*, 419−431.

(67) Wajon, J. E.; Rosenblatt, D. H.; Burrows, E. P. Oxidation of Phenol and Hydroquinone by Chlorine Dioxide. *Environ. Sci. Technol.* **1982**, *16* (7), 396−402.

(68) Glaze, W. H. Reaction products of ozone: A review. *Environ. Health Persp.* **1986**, *69*, 151−157.

(69) Dore, M.; Legube, B.; Langlais, B. Mecanisme de l'ozonation des herbicides derives de l'acide phenoxyacetique: 2−4D ET M.C.P.A. *Water Res.* **1980**, *14* (7), 767−773.

(70) Schuchmann, H.-P.; von Sonntag, C. Hydroxyl radical induced oxidation of diethyl ether in oxygenated aqeous solution. A product and pulse radiolysis study. *J. Phys. Chem.* **1982**, *86*, 1995−2000.

(71) EU drinking water directive. http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:31998L0083 (accessed September 12, 2017).

(72) Duguet, J. P.; Bernazeau, F.; Mallevialle, J. Research Note: Removal of Atrazine by Ozone and Ozone-Hydrogen Peroxide Combinations in Surface Water. *Ozone: Sci. Eng.* **1990**, *12* (2), 195−197.

(73) Galey, C.; Paslawski, D. Elimination des micropolluants par l̀ozone couplé avec le peroxyde d̀hydrogène dams le traitement de potabilisation de l̀eau. *L̀EAU, L̀INDUSTRIE, LES NUISANCES* **1993**, *Février 1993* (161), 46−49.

(74) Ministere de la Solidarité, d. l. S. e. d. l. P. S., Utilisation du peroxyde d'hydrogène pour le traitement des eaux destinées à la consumation humaine. In santé, D. g. d. l., Ed. Paris, 1990; Vol. *242*.

(75) Circulaire DG 5/VS 4 No 2000−166 du 28 mars 2000 relative aux produits de procédés de traitement des eaux destinées à la

consommation humaine. http://solidarites-sante.gouv.fr/fichiers/bo/2000/00-14/a0141021.htm (accessed September 19, 2017).

(76) Acero, J. L.; Stemmler, K.; von Gunten, U. Degradation kinetics of atrazine and its degradation products with ozone and OH radicals: A predictive tool for drinking water treatment. *Environ. Sci. Technol.* **2000**, *34*, 591−597.

(77) Acero, J. L.; Haderlein, S. B.; Schmidt, T. C.; von Gunten, U. MTBE Oxidation by Conventional Ozonation and the Combination Ozone/Hydrogen Peroxide:Efficiency of the Processes and Bromate Formation. *Environ. Sci. Technol.* **2001**, *35*, 4252−4259.

(78) Dowideit, P.; von Sonntag, C. Reactions of ozone with ethene and its methyl- and chlorine-substituted derivatives in aqueous solution. *Environ. Sci. Technol.* **1998**, *32*, 1112−1119.

(79) Muñoz, F.; von Sonntag, C. The reaction of ozone with tertiary amines including the complexing agents nitrilotriacetic acid (NTA) and ethylenediaminetetraacetic acid (EDTA) in aqueous solution. *J. Chem. Soc., Perkin Trans. 2* **2000**, 2029−2033.

(80) Leitzke, A.; Reisz, E.; Flyunt, R.; von Sonntag, C. The reaction of ozone with cinnamic acids: formation and decay of 2-hydroperoxy-2-hydroxyacetic acid. *J. Chem. Soc., Perkin Trans.2* **2001**, 793−797.

(81) Stefan, M. I.; Bolton, J. R. Mechanism of the Degradation of 1,4-Dioxane in Dilute Aqueous Solution Using the UV/Hydrogen Peroxide Process. *Environ. Sci. Technol.* **1998**, *32* (11), 1588−1595.

(82) Stefan, M. I.; Mack, J.; Boloton, J. R. Degradation Pathways during the Treatment of Methyl-tert-Butyl Ether by the UV/H2O2. *Environ. Sci. Technol.* **2000**, *34*, 650−658.

(83) Mons, M. N.; Heringa, M. B.; van Genderen, J.; Puijker, L. M.; Brand, W.; van Leeuwen, C. J.; Stoks, P.; van der Hoek, J. P.; van der Kooij, D. Use of the Threshold of Toxicological Concern (TTC) approach for deriving target values for drinking water contaminants. *Water Res.* **2013**, *47* (4), 1666−1678.

(84) von Sonntag, C. *Free-Radical-Induced DNA Damage and Its Repair. A Chemical Perspective*; Springer: Berlin, Heidelberg, 2006.

(85) von Sonntag, C.; Schuchmann, H.-P. Peroxyl radicals in aqueous solutions. In *Peroxyl radicals*, Alfassi, Z. B., Ed. John Wiley & Sons Ltd, 1997; pp 173−234.

(86) Hübner, U.; von Gunten, U.; Jekel, M. Evaluation of the persistence of transformation products from ozonation of trace organic compounds − A critical review. *Water Res.* **2015**, *68*, 150−170.

(87) Lee, Y.; von Gunten, U. Advances in predicting organic contaminant abatement during ozonation of municipal wastewater effluent: reaction kinetics, transformation products, and changes of biological effects. *Environmental Science: Water Research & Technology* **2016**, *2* (3), 421−442.

(88) Guo, X.; Minakata, D.; Niu, J.; Crittenden, J. Computer-Based First-Principles Kinetic Modeling of Degradation Pathways and Byproduct Fates in Aqueous-Phase Advanced Oxidation Processes. *Environ. Sci. Technol.* **2014**, *48* (10), 5718−5725.

(89) Lee, M.; Blum, L. C.; Schmid, E.; Fenner, K.; von Gunten, U. A computer-based prediction platform for the reaction of ozone with organic compounds in aqueous solution: kinetics and mechanisms. *Environmental Science: Processes & Impacts* **2017**, *19* (3), 465−476.

(90) Johnson, M. D.; Hornstein, B. J.; Wischnewsky, J., Ferrate(VI) Oxidation of Nitrogenous Compounds. In *Ferrates*; American Chemical Society, 2008; Vol. *985*, pp 177−188.

(91) Sharma, V. K. Oxidation of nitrogen-containing pollutants by novel ferrate(VI) technology: A review. *J. Environ. Sci. Health, Part A: Toxic/Hazard. Subst. Environ. Eng.* **2010**, *45* (6), 645−667.

(92) How, Z. T.; Linge, K. L.; Busetti, F.; Joll, C. A. Chlorination of Amino Acids: Reaction Pathways and Reaction Rates. *Environ. Sci. Technol.* **2017**, *51* (9), 4870−4876.

(93) Chen, Y.-T.; Chen, W.-R.; Liu, Z.-Q.; Lin, T.-F. Reaction Pathways and Kinetics of a Cyanobacterial Neurotoxin *β*-N-Methylamino-L-Alanine (BMAA) during Chlorination. *Environ. Sci. Technol.* **2017**, *51* (3), 1303−1311.

(94) Criquet, J.; Rodriguez, E. M.; Allard, S.; Wellauer, S.; Salhi, E.; Joll, C. A.; von Gunten, U. Reaction of bromine and chlorine with phenolic compounds and natural organic matter extracts − Electrophilic aromatic substitution and oxidation. *Water Res.* **2015**, *85*, 476−486.

(95) Vikesland, P. J.; Fiss, E. M.; Wigginton, K. R.; McNeill, K.; Arnold, W. A. Halogenation of Bisphenol-A, Triclosan, and Phenols in Chlorinated Waters Containing Iodide. *Environ. Sci. Technol.* **2013**, *47* (13), 6764−6772.

(96) Sivey, J. D.; Roberts, A. L. Assessing the Reactivity of Free Chlorine Constituents Cl2, Cl2O, and HOCl Toward Aromatic Ethers. *Environ. Sci. Technol.* **2012**, *46* (4), 2141−2147.

(97) Duirk, S. E.; Desetto, L. M.; Davis, G. M. Transformation of Organophosphorus Pesticides in the Presence of Aqueous Chlorine: Kinetics, Pathways, and Structure-Activity Relationships. *Environ. Sci. Technol.* **2009**, *43* (7), 2335−2340.

(98) Joo, S. H.; Mitch, W. A. Nitrile, Aldehyde, and Halonitroalkane Formation during Chlorination/Chloramination of Primary Amines. *Environ. Sci. Technol.* **2007**, *41* (4), 1288−1296.

(99) Dodd, M. C.; Huang, C. H. Aqueous chlorination of the antibacterial agent trimethoprim: Reaction kinetics and pathways. *Water Res.* **2007**, *41* (3), 647−655.

(100) Buth, J. M.; Arnold, W. A.; McNeill, K. Unexpected products and reaction mechanisms of the aqueous chlorination of cimetidine. *Environ. Sci. Technol.* **2007**, *41* (17), 6228−6233.

(101) Duirk, S. E.; Collette, T. W. Degradation of chlorpyrifos in aqueous chlorine solutions: Pathways, kinetics, and modeling. *Environ. Sci. Technol.* **2006**, *40* (2), 546−551.

(102) Bedner, M.; MacCrehan, W. A. Transformation of Acetaminophen by Chlorination Produces the Toxicants 1,4-Benzoquinone and N-Acetyl-p-benzoquinone Imine. *Environ. Sci. Technol.* **2006**, *40* (2), 516−522.

(103) Dodd, M. C.; Shah, A. D.; von Gunten, U.; Huang, C.-H. Interactions of Fluoroquinolone Antibacterial Agents with Aqueous Chlorine: Reaction Kinetics, Mechanisms, and Transformation Pathways. *Environ. Sci. Technol.* **2005**, *39* (18), 7065−7076.

(104) Rebenne, L. M.; Gonzalez, A. C.; Olson, T. M. Aqueous chlorination kinetics and mechanism of substituted dihydroxybenzenes. *Environ. Sci. Technol.* **1996**, *30* (7), 2235−2242.

(105) Rule, K. L.; Ebbett, V. R.; Vikesland, P. J. Formation of chloroform and chlorinated organics by free-chlorine-mediated oxidation of triclosan. *Environ. Sci. Technol.* **2005**, *39* (9), 3176−3185.

(106) Buth, J. M.; Steen, P. O.; Sueper, C.; Blumentritt, D.; Vikesland, P. J.; Arnold, W. A.; McNeill, K. Dioxin Photoproducts of Triclosan and Its Chlorinated Derivatives in Sediment Cores. *Environ. Sci. Technol.* **2010**, *44* (12), 4545−4551.

(107) Heeb, M. B.; Criquet, J.; Zimmermann-Steffens, S. G.; von Gunten, U. Oxidative treatment of bromide-containing waters: Formation of bromine and its reactions with inorganic and organic compounds — A critical review. *Water Res.* **2014**, *48*, 15−42.

(108) Acero, J. L.; Piriou, P.; von Gunten, U. Kinetics and mechanisms of formation of bromophenols during drinking water chlorination: Assessment of taste and odor development. *Water Res.* **2005**, *39* (13), 2979−2993.

(109) Rodriguez, E. M.; Acero, J. L.; Spoof, L.; Meriluoto, J. Oxidation of MC-LR and -RR with chlorine and potassium permanganate: Toxicity of the reaction products. *Water Res.* **2008**, *42* (6−7), 1744−1752.

(110) Willach, S.; Lutze, H. V.; Eckey, K.; Löppenberg, K.; Lüling, M.; Terhalle, J.; Wolbert, J.-B.; Jochmann, M. A.; Karst, U.; Schmidt, T. C. Degradation of sulfamethoxazole using ozone and chlorine dioxide - Compound-specific stable isotope analysis, transformation product analysis and mechanistic aspects. *Water Res.* **2017**, *122* (Supplement C), 280−289.

(111) Miao, H. F.; Han, H. H.; Ji, X. P.; Lu, M. F.; Huang, Z. X.; Ruan, W. Q. Oxidative transformation of oxcarbazepine by Cl_2, ClO_2 and O_3: characteristics and pathways. *Water Sci. Technol.: Water Supply* **2017**, *17* (1), 84.

(112) Wang, Y.; Liu, H.; Xie, Y.; Ni, T.; Liu, G. Oxidative removal of diclofenac by chlorine dioxide: Reaction kinetics and mechanism. *Chem. Eng. J.* **2015**, *279* (Supplement C), 409−415.

(113) Wang, P.; He, Y.-L.; Huang, C.-H. Oxidation of Antibiotic Agent Trimethoprim by Chlorine Dioxide: Reaction Kinetics and Pathways. *J. Environ. Eng.* **2012**, *138* (3), 360−366.

(114) Rav-Acha, C., Transformation of Aqueous Pollutants by Chlorine Dioxide: Reactions, Mechanisms and Products. In *Quality and Treatment of Drinking Water II*; Hrubec, J., Ed.; Springer Berlin Heidelberg: Berlin, Heidelberg, 1998; pp 143−175.

(115) Cwiertny, D. M.; Snyder, S. A.; Schlenk, D.; Kolodziej, E. P. Environmental Designer Drugs: When Transformation May Not Eliminate Risk. *Environ. Sci. Technol.* **2014**, *48* (20), 11737−11745.

(116) Dodd, M. C.; Kohler, H.-P. E.; von Gunten, U. Oxidation of Antibacterial Compounds by Ozone and Hydroxyl Radical: Elimination of Biological Activity during Aqueous Ozonation Processes. *Environ. Sci. Technol.* **2009**, *43* (7), 2498−2504.

(117) Dodd, M. C.; Rentsch, D.; Singer, H. P.; Kohler, H.-P. E.; von Gunten, U. Transformation of beta-Lactam Antibacterial Agents during Aqueous Ozonation: Reaction Pathways and Quantitative Bioassay of Biologically-Active Oxidation Products. *Environ. Sci. Technol.* **2010**, *44* (15), 5940−5948.

(118) Lange, F.; Cornelissen, S.; Kubac, D.; Sein, M. M.; von Sonntag, J.; Hannich, C. B.; Golloch, A.; Heipieper, H. J.; Moder, M.; von Sonntag, C. Degradation of macrolide antibiotics by ozone: A mechanistic case study with clarithromycin. *Chemosphere* **2006**, *65* (1), 17−23.

(119) Mestankova, H.; Schirmer, K.; Escher, B. I.; von Gunten, U.; Canonica, S. Removal of the antiviral agent oseltamivir and its biological activity by oxidative processes. *Environ. Pollut.* **2012**, *161*, 30−35.

(120) Keen, O. S.; Linden, K. G. Degradation of Antibiotic Activity during UV/H2O2 Advanced Oxidation and Photolysis in Wastewater Effluent. *Environ. Sci. Technol.* **2013**, *47* (22), 13020−13030.

(121) Suarez, S.; Dodd, M. C.; Omil, F.; von Gunten, U. Kinetics of triclosan oxidation by aqueous ozone and consequent loss of antibacterial activity: Relevance to municipal wastewater ozonation. *Water Res.* **2007**, *41* (12), 2481−2490.

(122) Huber, M. M.; Ternes, T. A.; von Gunten, U. Removal of estrogenic activity and formation of oxidation products during ozonation of 17 alpha-ethinylestradiol. *Environ. Sci. Technol.* **2004**, *38* (19), 5177−5186.

(123) Lee, Y.; Escher, B. I.; von Gunten, U. Efficient Removal of Estrogenic Activity during Oxidative Treatment of Waters Containing Steroid Estrogens. *Environ. Sci. Technol.* **2008**, *42* (17), 6333−6339.

(124) Linden, K. G.; Rosenfeldt, E. J.; Kullman, S. W. UV/H2O2 degradation of endocrine-disrupting chemicals in water evaluated via toxicity assays. *Water Sci. Technol.* **2007**, *55* (12), 313−319.

(125) Mestankova, H.; Escher, B.; Schirmer, K.; von Gunten, U.; Canonica, S. Evolution of algal toxicity during (photo)oxidative degradation of diuron. *Aquat. Toxicol.* **2011**, *101* (2), 466−473.

(126) Parker, A. M.; Lester, Y.; Spangler, E. K.; von Gunten, U.; Linden, K. G. UV/H2O2 advanced oxidation for abatement of organophosphorous pesticides and the effects on various toxicity screening assays. *Chemosphere* **2017**, *182*, 477−482.

(127) Fiss, E. M.; Rule, K. L.; Vikesland, P. J. Formation of Chloroform and Other Chlorinated Byproducts by Chlorination of Triclosan-Containing Antibacterial Products. *Environ. Sci. Technol.* **2007**, *41* (7), 2387−2394.

(128) Le Roux, J.; Gallard, H.; Croué, J.-P.; Papot, S.; Deborde, M. NDMA Formation by Chloramination of Ranitidine: Kinetics and Mechanism. *Environ. Sci. Technol.* **2012**, *46* (20), 11095−11103.

(129) Spahr, S.; Cirpka, O. A.; von Gunten, U.; Hofstetter, T. B. Formation of N-Nitrosodimethylamine during Chloramination of Secondary and Tertiary Amines: Role of Molecular Oxygen and Radical Intermediates. *Environ. Sci. Technol.* **2017**, *51* (1), 280−290.

(130) Schmidt, C. K.; Brauch, H.-J. N,N-Dimethylsulfamide as Precursor for N-Nitrosodimethylamine (NDMA) Formation upon Ozonation and its Fate During Drinking Water Treatment. *Environ. Sci. Technol.* **2008**, *42* (17), 6340−6346.

(131) von Gunten, U.; Salhi, E.; Schmidt, C. K.; Arnold, W. A. Kinetics and Mechanisms of N-Nitrosodimethylamine Formation upon Ozonation of N,N-Dimethylsulfamide-Containing Waters: Bromide Catalysis. *Environ. Sci. Technol.* **2010**, *44* (15), 5762−5768.

(132) Kosaka, K.; Asami, M.; Konno, Y.; Oya, M.; Kunikane, S. Identification of Antiyellowing Agents as Precursors of N-Nitrosodimethylamine Production on Ozonation from Sewage Treatment Plant Influent. *Environ. Sci. Technol.* **2009**, *43* (14), 5236−5241.

(133) Mestankova, H.; Parker, A. M.; Bramaz, N.; Canonica, S.; Schirmer, K.; von Gunten, U.; Linden, K. G. Transformation of Contaminant Candidate List (CCL3) compounds during ozonation and advanced oxidation processes in drinking water: Assessment of biological effects. *Water Res.* **2016**, *93*, 110−120.

(134) Leenheer, J. A.; Croue, J. P. Characterizing aquatic dissolved organic matter. *Environ. Sci. Technol.* **2003**, *37* (1), 18A−26A.

(135) Elovitz, M. S.; von Gunten, U.; Kaiser, H.-P., The influence of dissolved organic matter character on ozone decomposition rates and Rct. In *Natural Organic Matter and Disinfection by-Products, Characterization and Control in Drinking Water*; Barrett, S. E., Krasner, S. W., Amy, G., Eds.; American Chemical Society: Washington, DC., 2000; ACS Symposium Series 761.

(136) Korshin, G. V.; Li, C.-W.; Benjamin, M. M. Monitoring the properties of natural organic matter through UV spectroscopy: A consistent theory. *Water Res.* **1997**, *31* (7), 1787−1795.

(137) Aeschbacher, M.; Graf, C.; Schwarzenbach, R. P.; Sander, M. Antioxidant Properties of Humic Substances. *Environ. Sci. Technol.* **2012**, *46* (9), 4916−4925.

(138) Bourgin, M.; Beck, B.; Boehler, M.; Borowska, E.; Fleiner, J.; Salhi, E.; Teichler, R.; von Gunten, U.; Siegrist, H.; McArdell, C. S. Evaluation of a full-scale wastewater treatment plant upgraded with ozonation and biological post-treatments: Abatement of micropollutants, formation of transformation products and oxidation byproducts. *Water Res.* **2018**, *129*, 486−498.

(139) Hammes, F.; Salhi, E.; Koster, O.; Kaiser, H. P.; Egli, T.; von Gunten, U. Mechanistic and kinetic evaluation of organic disinfection by-product and assimilable organic carbon (AOC) formation during the ozonation of drinking water. *Water Res.* **2006**, *40* (12), 2275−2286.

(140) Wert, E. C.; Rosario-Ortiz, F. L.; Drury, D. D.; Snyder, S. A. Formation of oxidation byproducts from ozonation of wastewater. *Water Res.* **2007**, *41* (7), 1481−1490.

(141) Escher, B. I.; Bramaz, N.; Ort, C. JEM Spotlight: Monitoring the treatment efficiency of a full scale ozonation on a sewage treatment plant with a mode-of-action based test battery. *J. Environ. Monit.* **2009**, *11* (10), 1836−1846.

(142) Ramseier, M. K.; von Gunten, U. Mechanisms of Phenol Ozonation-Kinetics of Formation of Primary and Secondary Reaction Products. *Ozone: Sci. Eng.* **2009**, *31* (3), 201−215.

(143) Prasse, C.; Ford, B.; Nomura, D. K.; Sedlak, D. L. Unexpected transformation of dissolved phenols to toxic dicarbonyls by hadroxyl radicals and UV light. *Proc. Natl. Acad. Sci. U. S. A.* **2018**, *115*, 2311.

(144) Tentscher, P. R.; Bourgin, M.; von Gunten, U., Ozonation of para-substituted phenolic compounds yields p-benzoquinones, other alpha, beta.unsaturated ketones, and substituted catechols. *Environ. Sci. Technol.* **2018**, *in press*, http://dx.doi.org/10.1021/acs.est.8b00011.10.1021/acs.est.8b00011

(145) Prasse, C.; Stalter, D.; Schulte-Oehlmann, U.; Oehlmann, J.; Ternes, T. A. Spoilt for choice: A critical review on the chemical and biological assessment of current wastewater treatment technologies. *Water Res.* **2015**, *87*, 237−270.

(146) Stalter, D.; Magdeburg, A.; Weil, M.; Knacker, T.; Oehlmann, J. Toxication or detoxication? In vivo toxicity assessment of ozonation as advanced wastewater treatment with the rainbow trout. *Water Res.* **2010**, *44* (2), 439−448.

(147) Magdeburg, A.; Stalter, D.; Schlüsener, M.; Ternes, T.; Oehlmann, J. Evaluating the efficiency of advanced wastewater treatment: Target analysis of organic contaminants and (geno-)toxicity assessment tell a different story. *Water Res.* **2014**, *50* (0), 35−47.

(148) Hollender, J.; Zimmermann, S. G.; Koepke, S.; Krauss, M.; McArdell, C. S.; Ort, C.; Singer, H.; von Gunten, U.; Siegrist, H. 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* (20), 7862−7869.

(149) Zimmermann, S. G.; Wittenwiler, M.; Hollender, J.; Krauss, M.; Ort, C.; Siegrist, H.; von Gunten, U. Kinetic assessment and modeling of an ozonation step for full-scale municipal wastewater treatment: Micropollutant oxidation, by-product formation and disinfection. *Water Res.* **2011**, *45* (2), 605−617.

(150) Gerrity, D.; Gamage, S.; Holady, J. C.; Mawhinney, D. B.; Quiones, O.; Trenholm, R. A.; Snyder, S. A. Pilot-scale evaluation of ozone and biological activated carbon for trace organic contaminant mitigation and disinfection. *Water Res.* **2011**, *45* (5), 2155−2165.

(151) Plewa, M. J.; Muellner, M. G.; Richardson, S. D.; Fasano, F.; Buettner, K. M.; Woo, Y.-T.; McKague, A. B.; Wagner, E. D. Occurrence, Synthesis, and Mammalian Cell Cytotoxicity and Genotoxicity of Haloacetamides: An Emerging Class of Nitrogenous Drinking Water Disinfection Byproducts. *Environ. Sci. Technol.* **2008**, *42* (3), 955−961.

(152) Plewa, M. J.; Wagner, E. D.; Richardson, S. D.; Thruston, A. D.; Woo, Y.-T.; McKague, A. B. Chemical and Biological Characterization of Newly Discovered Iodoacid Drinking Water Disinfection Byproducts. *Environ. Sci. Technol.* **2004**, *38* (18), 4713−4722.

(153) von Gunten, U.; Hoigné, J. Bromate formation during ozonation of bromide-containing waters: Interaction of ozone and hydroxyl radical reactions. *Environ. Sci. Technol.* **1994**, *28*, 1234−1242.

(154) Pinkernell, U.; von Gunten, U. Bromate minimization during ozonation: mechanistic considerations. *Environ. Sci. Technol.* **2001**, *35*, 2525−2531.

(155) Soltermann, F.; Abegglen, C.; Tschui, M.; Stahel, S.; von Gunten, U. Options and limitations for bromate control during ozonation of wastewater. *Water Res.* **2017**, *116*, 76−85.

(156) Song, R.; Donohoe, C.; Minear, R.; Westerhoff, P.; Ozekin, K.; Amy, G. Empirical Modeling of Bromate Formation during ozonation of Bromide-Containing Waters. *Water Res.* **1996**, *30*, 1161−1168.

(157) von Gunten, U.; Oliveras, Y. Advanced Oxidation of bromide-containing waters: bromate formation mechanisms. *Environ. Sci. Technol.* **1998**, *32*, 63−70.

(158) von Gunten, U.; Oliveras, Y. Kinetics of the reaction between hydrogen peroxide and hypobromous acid: Implication on water treatment and natural systems. *Water Res.* **1997**, *31*, 900−906.

(159) Krasner, S. W.; Glaze, W. H.; Weinberg, H. S.; Daniel, P. A.; Najm, I. N. Formation and Control of Bromate During Ozonation of Waters Containing Bromide. *J. - Am. Water Works Assoc.* **1993**, *85* (1), 73−81.

(160) Fischbacher, A.; Löppenberg, K.; von Sonntag, C.; Schmidt, T. C. A New Reaction Pathway for Bromite to Bromate in the Ozonation of Bromide. *Environ. Sci. Technol.* **2015**, *49* (19), 11714−11720.

(161) Siddiqui, M.; Amy, G.; Ozekin, K.; Zhai, W.; Westerhoff, P. Alternative strategies for removing bromate. *J. - Am. Water Works Assoc.* **1994**, *86* (10), 81−96.

(162) Kirisits, M. J.; Snoeyink, V. L.; Kruithof, J. C. The reduction of bromate by granular activated carbon. *Water Res.* **2000**, *34* (17), 4250−4260.

(163) Buffle, M. O.; Galli, S.; von Gunten, U. Enhanced bromate control during ozonation: The chlorine-ammonia process. *Environ. Sci. Technol.* **2004**, *38* (19), 5187−5195.

(164) Neemann, J.; Hulsey, R.; Rexing, D.; Wert, E. Controlling bromate formation during ozonation with chlorine and ammonia. *J. - Am. Water Works Assoc.* **2004**, *96* (2), 26−29.

(165) Symons, J. M.; Zheng, M. C. H. Technical note: Does hydroxyl radical oxidize bromide to bromate? *J. - Am. Water Works Assoc.* **1997**, *89* (6), 106−109.

(166) Rosenfeldt, E. J.; Linden, K. G.; Canonica, S.; von Gunten, U. Comparison of the efficiency of (OH)-O-center dot radical formation during ozonation and the advanced oxidation Processes O-3/H2O2 and UV/H2O2 (vol 40, pg 20, 2006). *Water Res.* **2008**, *42* (10−11), 2836−2838.

(167) Katsoyiannis, I. A.; Canonica, S.; von Gunten, U. Efficiency and energy requirements for the transformation of organic micropollutants by ozone, O3/H2O2 and UV/H2O2. *Water Res.* **2011**, *45* (13), 3811−3822.

(168) Lee, Y.; Gerrity, D.; Lee, M.; Gamage, S.; Pisarenko, A.; Trenholm, R. A.; Canonica, S.; Snyder, S. A.; von Gunten, U. Organic Contaminant Abatement in Reclaimed Water by UV/H2O2 and a Combined Process Consisting of O3/H2O2 Followed by UV/H2O2: Prediction of Abatement Efficiency, Energy Consumption, and Byproduct Formation. *Environ. Sci. Technol.* **2016**, *50* (7), 3809−3819.

(169) Staehelin, J.; Hoigné, J. Decomposition of ozone in water: rate of initiation by hydroxide ions and hydrogen peroxide. *Environ. Sci. Technol.* **1982**, *16*, 676−681.

(170) EU project Neptune. http://www.eu-neptune.org/index_EN (accessed September 19, 2017),.

(171) Poseidon EU project Poseidon. http://undine.bafg.de/servlet/is/2884/index3322.html?lang=de.

(172) EU Project DEMAU: Demonstration of promising technologies to address emerging pollutants in water and wastewater. http://demeau-fp7.eu/ (accessed September 20, 2017),.

(173) Eggen, R. I. L.; Hollender, J.; Joss, A.; Schärer, M.; Stamm, C. Reducing the Discharge of Micropollutants in the Aquatic Environment: The Benefits of Upgrading Wastewater Treatment Plants. *Environ. Sci. Technol.* **2014**, *48* (14), 7683−7689.

(174) Stamm, C.; Eggen, R. I. L.; Hering, J. G.; Hollender, J.; Joss, A.; Schärer, M. Micropollutant Removal from Wastewater: Facts and Decision-Making Despite Uncertainty. *Environ. Sci. Technol.* **2015**, *49* (11), 6374−6375.

(175) Joss, A.; Siegrist, H.; Ternes, T. A. Are we about to upgrade wastewater treatment for removing organic micropollutants? *Water Sci. Technol.* **2008**, *57* (2), 251−255.

(176) Ort, C.; Hollender, J.; Schaerer, M.; Siegrist, H. Model-Based Evaluation of Reduction Strategies for Micropollutants from Wastewater Treatment Plants in Complex River Networks. *Environ. Sci. Technol.* **2009**, *43* (9), 3214−3220.

(177) Abegglen, C.; Böhler, M.; Hollender, J.; Zimmermann, S. G.; Zwickenpflug, B.; von Gunten, U.; Siegrist, H.; Thonney, D. Mikroverunreinigungen in Kläranlagen. Technische Verfahren zur Elimination von organischen Spurenstoffen. *Gas Wasser Abwasser* **2010**, No. 7, 587−594.

(178) Ternes, T. A. Occurrence of drugs in German sewage treatment plants and rivers. *Water Res.* **1998**, *32*, 3245−3260.

(179) Ternes, T. A.; Joss, A.; Siegrist, H. Peer Reviewed: Scrutinizing Pharmaceuticals and Personal Care Products in Wastewater Treatment. *Environ. Sci. Technol.* **2004**, *38* (20), 392A−399A.

(180) Ternes, T.; Joss, A. *Human Pharmaceuticals, Hormones and Fragrances: The Challenge of Micropollutants in Urban Water Management*; IWA: London, 2006.

(181) Snyder, S. A.; Westerhoff, P.; Yoon, Y.; Sedlak, D. L. Pharmaceuticals, personal care products, and endocrine disruptors in water: Implications for the water industry. *Environ. Eng. Sci.* **2003**, *20* (5), 449−469.

(182) Snyder, S. A. Occurrence, Treatment, and Toxicological Relevance of EDCs and Pharmaceuticals in Water. *Ozone: Sci. Eng.* **2008**, *30* (1), 65−69.

(183) Tyler, C. R.; Jobling, S.; Sumpter, J. P. Endocrine disruption in wildlife: A critical review of the evidence. *Crit. Rev. Toxicol.* **1998**, *28* (4), 319−361.

(184) Sumpter, J. P.; Johnson, A. C. 10th Anniversary Perspective: Reflections on endocrine disruption in the aquatic environment: from known knowns to unknown unknowns (and many things in between). *J. Environ. Monit.* **2008**, *10* (12), 1476−1485.

(185) Sumpter, J. P.; Jobling, S. Vitellogenesis as a biomarker for estrogenic contamination of the aquatic environment. *Environ. Health Perspect.* **1995**, *103* (Suppl 7), 173−178.

(186) Jobling, S.; Nolan, M.; Tyler, C. R.; Brighty, G.; Sumpter, J. P. Widespread Sexual Disruption in Wild Fish. *Environ. Sci. Technol.* **1998**, *32* (17), 2498−2506.

(187) Kidd, K. A.; Blanchfield, P. J.; Mills, K. H.; Palace, V. P.; Evans, R. E.; Lazorchak, J. M.; Flick, R. W. Collapse of a fish population after

exposure to a synthetic estrogen. *Proc. Natl. Acad. Sci. U. S. A.* **2007**, *104* (21), 8897−8901.

(188) Ternes, T. A.; Prasse, C.; Eversloh, C. L.; Knopp, G.; Cornel, P.; Schulte-Oehlmann, U.; Schwartz, T.; Alexander, J.; Seitz, W.; Coors, A.; Oehlmann, J. Integrated Evaluation Concept to Assess the Efficacy of Advanced Wastewater Treatment Processes for the Elimination of Micropollutants and Pathogens. *Environ. Sci. Technol.* **2017**, *51* (1), 308−319.

(189) Ternes, T. A.; Meisenheimer, M.; McDowell, D.; Sacher, F.; Brauch, H. J.; Haist-Gulde, B.; Preuss, G.; Wilme, U.; Zulei-Seibert, N. Removal of pharmaceuticals during drinking water treatment. *Environ. Sci. Technol.* **2002**, *36* (17), 3855−3863.

(190) Besnault, S.; Ruel, M. S.; Baig, S.; Esperanza, M.; Budzinski, H. In *Technical, ecomomic and environmental evaluation of advanced tertiary treatments for micropollutants removal (oxidation and adsorption)*, ECOSTP, Verona, Italy, 2014; IWA: Verona, Italy, 2014; pp 262−265.

(191) Lloyd Owen, D. A. Public−private partnerships in the water reuse sector: a global assessment. *International Journal of Water Resources Development* **2016**, *32* (4), 526−535.

(192) Horne, J. Policy issues confronting Australian urban water reuse. *International Journal of Water Resources Development* **2016**, *32* (4), 573−589.

(193) Zodrow, K. R.; Li, Q.; Buono, R. M.; Chen, W.; Daigger, G.; Dueñas-Osorio, L.; Elimelech, M.; Huang, X.; Jiang, G.; Kim, J.-H.; Logan, B. E.; Sedlak, D. L.; Westerhoff, P.; Alvarez, P. J. J. Advanced Materials, Technologies, and Complex Systems Analyses: Emerging Opportunities to Enhance Urban Water Security. *Environ. Sci. Technol.* **2017**, *51* (18), 10274−10281.

(194) Olivieri, A. W.; Crook, J.; Anderson, M. A.; Bull, R. J.; Drewes, J. E.; Haas, C.; Jakubowski, W.; McCarty, P. L.; Nelson, K. L.; Rose, J. B.; Sedlak, D. L.; Wade, T. J. *Evaluation of the Feasibility of Developing Uniform Water Recycling Criteria for Direct Potable Reuse*; National Water Research Institute: Fountain Valley, 2016.

(195) Harris-Lovett, S.; Sedlak, D. *The History of Water Reuse in California*; University California Press: Oakland, 2015; p 220−243.

(196) Trussell, R. R.; Anderson, H. A.; Archuleta, E. G.; Crook, J.; Drewes, J. E.; Fort, D. D.; Haas, C.; Haddad, B. M.; Huggett, D. B.; Jiang, S.; Sedlak, D. L.; Snyder, S.; Whittaker, M. H.; Whittington, D. *Water Reuse: Potential for Expanding the Nation̓s Water Supply Through Reuse of Municipal Wastewater*; 978−0−309−25749−7; The National Academies Press: Washington, DC, 2012; p 276.

(197) Leverenz, H. L.; Tchobanoglous, G.; Asano, T. Direct potable reuse: a future imperative. *J. Water Reuse Desalin.* **2011**, *1* (1), 2.

(198) Australian guidelines for water recycling: Managing health and environmental risks - stromwater harvesting and reuse. http://www.agriculture.gov.au/water/quality/nwqms/nwqms-australian-guidelines-water-recycling-managing-health-phase1 (accessed September 27, 2017),.

(199) Barazesh, J. M.; Prasse, C.; Wenk, J.; Berg, S.; Remucal, C. K.; Sedlak, D. L. Trace Element Removal in Distributed Drinking Water Treatment Systems by Cathodic H2O2 Production and UV Photolysis. *Environ. Sci. Technol.* **2018**, *52* (1), 195−204.

(200) Barazesh, J. M.; Prasse, C.; Sedlak, D. L. Electrochemical Transformation of Trace Organic Contaminants in the Presence of Halide and Carbonate Ions. *Environ. Sci. Technol.* **2016**, *50* (18), 10143−10152.

(201) Barazesh, J. M.; Hennebel, T.; Jasper, J. T.; Sedlak, D. L. Modular Advanced Oxidation Process Enabled by Cathodic Hydrogen Peroxide Production. *Environ. Sci. Technol.* **2015**, *49* (12), 7391−7399.

(202) Schymanski, E. L.; Singer, H. P.; Longrée, P.; Loos, M.; Ruff, M.; Stravs, M. A.; Ripollés Vidal, C.; Hollender, J. Strategies to Characterize Polar Organic Contamination in Wastewater: Exploring the Capability of High Resolution Mass Spectrometry. *Environ. Sci. Technol.* **2014**, *48* (3), 1811−1818.

(203) Robles-Molina, J.; Lara-Ortega, F. J.; Gilbert-López, B.; García-Reyes, J. F.; Molina-Díaz, A. Multi-residue method for the determination of over 400 priority and emerging pollutants in water and wastewater by solid-phase extraction and liquid chromatography-time-of-flight mass spectrometry. *Journal of Chromatography A* **2014**, *1350* (SupplementC), 30−43.

(204) Bletsou, A. A.; Jeon, J.; Hollender, J.; Archontaki, E.; Thomaidis, N. S. Targeted and non-targeted liquid chromatography-mass spectrometric workflows for identification of transformation products of emerging pollutants in the aquatic environment. *TrAC, Trends Anal. Chem.* **2015**, *66* (Supplement C), 32−44.

(205) NDRL/NIST Solution Kinetics Database on the Web. http://kinetics.nist.gov/solution/ (accessed September 26, 2017),.

(206) Hoigné, J.; Bader, H. Kinetics of reactions of chlorine dioxide (OClO) in water - I. Rate constants for inorganic and organic compounds. *Water Res.* **1994**, *28*, 45−55.

(207) Tratnyek, P. G.; Hoigne, J. Oxidation of substituted phenols in the environment: a QSAR analysis of rate constants for reaction with singlet oxygen. *Environ. Sci. Technol.* **1991**, *25* (9), 1596−1604.

(208) Lee, Y.; von Gunten, U. Quantitative structure-Activity relationships (QSARs) for the transformation of organic micropollutants during oxidative water treatment. *Water Res.* **2012**, *46* (19), 6177−6195.

(209) Lee, M.; Zimmermann-Steffens, S. G.; Arey, J. S.; Fenner, K.; von Gunten, U. Development of Prediction Models for the Reactivity of Organic Compounds with Ozone in Aqueous Solution by Quantum Chemical Calculations: The Role of Delocalized and Localized Molecular Orbitals. *Environ. Sci. Technol.* **2015**, *49* (16), 9925−9935.

(210) Minakata, D.; Crittenden, J. Linear Free Energy Relationships between Aqueous phase Hydroxyl Radical Reaction Rate Constants and Free Energy of Activation. *Environ. Sci. Technol.* **2011**, *45* (8), 3479−3486.

(211) Minakata, D.; Li, K.; Westerhoff, P.; Crittenden, J. Development of a Group Contribution Method To Predict Aqueous Phase Hydroxyl Radical (HO center dot) Reaction Rate Constants. *Environ. Sci. Technol.* **2009**, *43* (16), 6220−6227.

(212) Borowska, E.; Bourgin, M.; Hollender, J.; Kienle, C.; McArdell, C. S.; von Gunten, U. Oxidation of cetirizine, fexofenadine and hydrochlorothiazide during ozonation: Kinetics and formation of transformation products. *Water Res.* **2016**, *94*, 350−362.

(213) Tekle-Röttering, A.; von Sonntag, C.; Reisz, E.; Eyser, C. v.; Lutze, H. V.; Türk, J.; Naumov, S.; Schmidt, W.; Schmidt, T. C. Ozonation of anilines: Kinetics, stoichiometry, product identification and elucidation of pathways. *Water Res.* **2016**, *98*, 147−159.

(214) Tekle-Röttering, A.; Reisz, E.; Jewell, K. S.; Lutze, H. V.; Ternes, T. A.; Schmidt, W.; Schmidt, T. C. Ozonation of pyridine and other N-heterocyclic aromatic compounds: Kinetics, stoichiometry, identification of products and elucidation of pathways. *Water Res.* **2016**, *102*, 582−593.

(215) Tekle-Röttering, A.; Jewell, K. S.; Reisz, E.; Lutze, H. V.; Ternes, T. A.; Schmidt, W.; Schmidt, T. C. Ozonation of piperidine, piperazine and morpholine: Kinetics, stoichiometry, product formation and mechanistic considerations. *Water Res.* **2016**, *88*, 960−971.

(216) Radjenovic, J.; Godehardt, M.; Petrovic, M.; Hein, A.; Farré, M.; Jekel, M.; Barcelo, D. Evidencing Generation of Persistent Ozonation Products of Antibiotics Roxithromycin and Trimethoprim. *Environ. Sci. Technol.* **2009**, *43* (17), 6808−6815.

(217) Sein, M. M.; Zedda, M.; Tuerk, J.; Schmidt, T. C.; Golloch, A.; von Sonntag, C. Oxidation of Diclofenac with Ozone in Aqueous Solution. *Environ. Sci. Technol.* **2008**, *42* (17), 6656−6662.

(218) Mvula, E.; von Sonntag, C. Ozonolysis of phenols in aqueous solution. *Org. Biomol. Chem.* **2003**, *1* (10), 1749−1756.

(219) Leitzke, A.; Flyunt, R.; Theruvathu, J. A.; von Sonntag, C. Ozonolysis of vinyl compounds, $CH_2{=}CH\text{-}X$, in aqueous solution - the chemistries of the ensuing formyl compounds and hydroperoxides. *Org. Biomol. Chem.* **2003**, *1* (6), 1012−1019.

(220) vonSonntag, C. Degradation of aromatics by Advanced Oxidation Processes in water remediation: Some basic considerations. *Journal of Water Supply Research and Technology-Aqua* **1996**, *45* (2), 84−91.

(221) Zimmermann, S. G.; Schmukat, A.; Schulz, M.; Benner, J.; von Gunten, U.; Ternes, T. A. Kinetic and Mechanistic Investigations of

the Oxidation of Tramadol by Ferrate and Ozone. *Environ. Sci. Technol.* **2012**, *46* (2), 876−884.
(222) Prasse, C.; Wagner, M.; Schulz, R.; Ternes, T. A. Oxidation of the Antiviral Drug Acyclovir and Its Biodegradation Product Carboxy-acyclovir with Ozone: Kinetics and Identification of Oxidation Products. *Environ. Sci. Technol.* **2012**, *46* (4), 2169−2178.
(223) Benner, J.; Ternes, T. A. Ozonation of Metoprolol: Elucidation of Oxidation Pathways and Major Oxidation Products. *Environ. Sci. Technol.* **2009**, *43* (14), 5472−5480.
(224) Benner, J.; Ternes, T. A. Ozonation of Propranolol: Formation of Oxidation Products. *Environ. Sci. Technol.* **2009**, *43* (13), 5086−5093.
(225) Escher, B. I.; Baumgartner, R.; Koller, M.; Treyer, K.; Lienert, J.; McArdell, C. S. Environmental toxicology and risk assessment of pharmaceuticals from hospital wastewater. *Water Res.* **2011**, *45* (1), 75−92.
(226) Kern, S.; Fenner, K.; Singer, H. P.; Schwarzenbach, R. P.; Hollender, J. Identification of Transformation Products of Organic Contaminants in Natural Waters by Computer-Aided Prediction and High-Resolution Mass Spectrometry. *Environ. Sci. Technol.* **2009**, *43* (18), 7039−7046.
(227) Howard, P. H.; Stiteler, W. M.; Meylan, W. M.; Hueber, A. E.; Beauman, J. A.; Larosche, M. E.; Boethling, R. S. Predictive model for aerobic biodegradability developed from a file of evaluated biodegradation data. *Environ. Toxicol. Chem.* **1992**, *11* (5), 593−603.
(228) Latino, D. A. R. S.; Wicker, J.; Gutlein, M.; Schmid, E.; Kramer, S.; Fenner, K. Eawag-Soil in enviPath: a new resource for exploring regulatory pesticide soil biodegradation pathways and half-life data. *Environmental Science: Processes & Impacts* **2017**, *19* (3), 449−464.
(229) Schollée, J. E.; Bourgin, M.; von Gunten, U.; McArdell, C. S.; Hollender, J. Non-target screening to trace ozonation transformation products in a wastewater treatment train including different post-treatments. *Water Res.* **2018**, submitted.
(230) Gold, L. S. The Carcinogenic Potency Database (CPDB). https://toxnet.nlm.nih.gov/cpdb/.
(231) Benigni, R.; Bossa, C. Mechanisms of Chemical Carcinogenicity and Mutagenicity: A Review with Implications for Predictive Toxicology. *Chem. Rev.* **2011**, *111* (4), 2507−2536.
(232) Toxtree - Toxic Hazard Estimation by Decision Tree Approach. http://toxtree.sourceforge.net/.
(233) QSAR Toolbox. http://www.qsartoolbox.org.
(234) Online Chemical Database. https://ochem.eu/home/show.do.
(235) Radjenovic, J.; Sedlak, D. L. Challenges and Opportunities for Electrochemical Processes as Next-Generation Technologies for the Treatment of Contaminated Water. *Environ. Sci. Technol.* **2015**, *49* (19), 11292−11302.
(236) Bakheet, B.; Yuan, S.; Li, Z.; Wang, H.; Zuo, J.; Komarneni, S.; Wang, Y. Electro-peroxone treatment of Orange II dye wastewater. *Water Res.* **2013**, *47* (16), 6234−6243.
(237) Da Silva, L. M.; De Faria, L. A.; Boodts, J. F. C. Green processes for environmental application. Electrochemical ozone production. *Pure Appl. Chem.* **2001**, *73* (12), 1871−1884.
(238) Legube, B.; Karpel Vel Leitner, N. Catalytic ozonation: a promising advanced oxidation technology for water treatment. *Catal. Today* **1999**, *53* (1), 61−72.
(239) Li, W.; Lu, X.; Xu, K.; Qu, J.; Qiang, Z. Cerium incorporated MCM-48 (Ce-MCM-48) as a catalyst to inhibit bromate formation during ozonation of bromide-containing water: Efficacy and mechanism. *Water Res.* **2015**, *86*, 2−8.
(240) Fischbacher, A.; von Sonntag, J.; von Sonntag, C.; Schmidt, T. C. The $^{\bullet}$OH Radical Yield in the H_2O_2 + O_3 (Peroxone) Reaction. *Environ. Sci. Technol.* **2013**, *47* (17), 9959−9964.
(241) Merle, T.; Pronk, W.; von Gunten, U. MEMBRO3X, a Novel Combination of a Membrane Contactor with Advanced Oxidation (O_3/H_2O_2) for Simultaneous Micropollutant Abatement and Bromate Minimization. *Environ. Sci. Technol. Lett.* **2017**, *4* (5), 180−185.
(242) Bowman, R. H., Hipox advanced oxidation of TBA and MTBE in groundwater. In *Contaminated Soils, Sediments and Water: Science in the Real World, Vol 9*; Calabrese, E. J.; Kostecki, P. T.; Dragun, J., Eds.; Springer: New York, 2005; Vol. *9*, pp 299−313.
(243) Knol, A. H.; Lekkerkerker-Teunissen, K.; Houtman, C. J.; Scheideler, J.; Ried, A.; van Dijk, J. C. Conversion of organic micropollutants with limited bromate formation during the Peroxone process in drinking water treatment. *Drinking Water Eng. Sci.* **2015**, *8* (2), 25−34.
(244) Yao, W.; Qu, Q.; von Gunten, U.; Chen, C.; Yu, G.; Wang, Y. Comparison of methylisoborneol and geosmin abatement in surface water by conventional ozonation and an electro-peroxone process. *Water Res.* **2017**, *108*, 373−382.
(245) Alapi, T.; Schrantz, K. A.; Arany, E.; Kozmer, Z., Vacuum UV radiation-driven processes. In *Advanced Oxidation Processes for Water Treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publishing: London, 2018.
(246) Zoschke, K.; Börnick, H.; Worch, E. Vacuum-UV radiation at 185 nm in water treatment − A review. *Water Res.* **2014**, *52* (0), 131−145.
(247) Bagheri, M.; Mohseni, M. A study of enhanced performance of VUV/UV process for the degradation of micropollutants from contaminated water. *J. Hazard. Mater.* **2015**, *294* (Supplement C), 1−8.
(248) Remucal, C. K.; Manley, D. Emerging investigators series: the efficacy of chlorine photolysis as an advanced oxidation process for drinking water treatment. *Environmental Science: Water Research & Technology* **2016**, *2* (4), 565−579.
(249) Nowell, L. H.; Hoigné, J. Photolysis of Aqueous chlorine at sunligth and ultraviolet wavelengths-II Hydroxyl radical Production. *Water Res.* **1992**, *26*, 599−605.
(250) Watts, M. J.; Linden, K. G. Chlorine photolysis and subsequent OH radical production during UV treatment of chlorinated water. *Water Res.* **2007**, *41* (13), 2871−2878.
(251) Zhou, P.; Di Giovanni, G. D.; Meschke, J. S.; Dodd, M. C. Enhanced Inactivation of Cryptosporidium parvum Oocysts during Solar Photolysis of Free Available Chlorine. *Environ. Sci. Technol. Lett.* **2014**, *1* (11), 453−458.
(252) De Laat, J.; Stefan, M. I., UV/Chlorine process. In *Advanced Oxidation Processes for Water Treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publishing: London, 2018.
(253) Wang, D.; Bolton, J. R.; Andrews, S. A.; Hofmann, R. Formation of disinfection by-products in the ultraviolet/chlorine advanced oxidation process. *Sci. Total Environ.* **2015**, *518*, 49−57.
(254) Fang, J.; Zhao, Q.; Fan, C.; Shang, C.; Fu, Y.; Zhang, X. Bromate formation from the oxidation of bromide in the UV/chlorine process with low pressure and medium pressure UV lamps. *Chemosphere* **2017**, *183*, 582−588.
(255) Chuang, Y.-H.; Chen, S.; Chinn, C. J.; Mitch, W. A. Comparing the UV/Monochloramine and UV/Free Chlorine Advanced Oxidation Processes (AOPs) to the UV/Hydrogen Peroxide AOP Under Scenarios Relevant to Potable Reuse. *Environ. Sci. Technol.* **2017**, *51* (23), 13859−13868.
(256) Patton, S.; Li, W.; Couch, K. D.; Mezyk, S. P.; Ishida, K. P.; Liu, H. Impact of the Ultraviolet Photolysis of Monochloramine on 1,4-Dioxane Removal: New Insights into Potable Water Reuse. *Environ. Sci. Technol. Lett.* **2017**, *4* (1), 26−30.
(257) Beck, S. E.; Ryu, H.; Boczek, L. A.; Cashdollar, J. L.; Jeanis, K. M.; Rosenblum, J. S.; Lawal, O. R.; Linden, K. G. Evaluating UV-C LED disinfection performance and investigating potential dual-wavelength synergy. *Water Res.* **2017**, *109* (Supplement C), 207−216.
(258) Chen, J.; Loeb, S.; Kim, J.-H. LED revolution: fundamentals and prospects for UV disinfection applications. *Environmental Science: Water Research & Technology* **2017**, *3* (2), 188−202.
(259) Loeb, S.; Hofmann, R.; Kim, J.-H. Beyond the Pipeline: Assessing the Efficiency Limits of Advanced Technologies for Solar Water Disinfection. *Environ. Sci. Technol. Lett.* **2016**, *3* (3), 73−80.
(260) Song, K.; Mohseni, M.; Taghipour, F. Application of ultraviolet light-emitting diodes (UV-LEDs) for water disinfection: A review. *Water Res.* **2016**, *94* (Supplement C), 341−349.

DOI: 10.1021/acs.est.8b00586
Environ. Sci. Technol. 2018, 52, 5062−5075

(261) Harris, T. R.; Pagan, J.; Batoni, P. Optical and Fluidic Co-Design of a UV-LED Water Disinfection Chamber. *ECS Trans.* **2013**, *45* (17), 11−18.

(262) Kneissl, M. UV-LED efficiency. http://www.ifkp.tu-berlin.de/fileadmin/i1/Kneissl/UV_LED_Efficiency_AUG_2017.pdf (22.11.).

(263) Stefan, M. I., UV/hydrogen peroxide process. In *Advanced Oxidation Processes for Water Treatment: Fundamentals and Applications*;Stefan, M. I., Ed.; IWA Publishing: London, 2018.

(264) Yoshihiko, M.; Masahiro, K.; Suguru, N. Development and future of ultraviolet light-emitting diodes: UV-LED will replace the UV lamp. *Semicond. Sci. Technol.* **2014**, *29* (8), 084004.

(265) Haitz, R.; Tsao, J. Y. Solid-state lighting: 'The case' 10 years after and future prospects. *Phys. Status Solidi A* **2011**, *208* (1), 17−29.

(266) Feng, Y.; Yang, L.; Liu, J.; Logan, B. E. Electrochemical technologies for wastewater treatment and resource reclamation. *Environmental Science: Water Research & Technology* **2016**, *2* (5), 800−831.

(267) Oturan, M. A.; Aaron, J.-J. Advanced Oxidation Processes in Water/Wastewater Treatment: Principles and Applications. A Review. *Crit. Rev. Environ. Sci. Technol.* **2014**, *44* (23), 2577−2641.

(268) Vecitis, C. D.; Park, H.; Cheng, J.; Mader, B.; Hoffmann, M. Treatment technologies for aqueous perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA). *Front. Environ. Sci. Eng. China* **2009**, *3* (2), 129−151.

(269) Foster, J. E. Plasma-based water purification: Challenges and prospects for the future. *Phys. Plasmas* **2017**, *24* (5), 055501.

(270) Gerrity, D.; Stanford, B. D.; Trenholm, R. A.; Snyder, S. A. An evaluation of a pilot-scale nonthermal plasma advanced oxidation process for trace organic compound degradation. *Water Res.* **2010**, *44* (2), 493−504.

(271) Thagard, S. M.; Locke, B. R., Electrical discharge plasma for water treatment. In *Advanced Oxidation Processes for Water Treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publishing: London, 2018.

(272) Larpparisudthi, O. A.; Mason, T. J.; Paniwnyk, L., Ultrasound wave-based AOPs. In *Advanced Oxidation Processes for Water Treatment: Fundamentals and Applications*; Stefan, M. I., Ed.; IWA Publishing: London, 2018.

(273) Kim, M. H.; Cho, J. H.; Park, S. J.; Eden, J. G. Modular and efficient ozone systems based on massively parallel chemical processing in microchannel plasma arrays: performance and commercialization. *Eur. Phys. J.: Spec. Top.* **2017**, *226* (13), 2923−2944.

(274) Tang, L. L.; DeNardo, M. A.; Gayathri, C.; Gil, R. R.; Kanda, R.; Collins, T. J. TAML/H2O2 Oxidative Degradation of Metaldehyde: Pursuing Better Water Treatment for the Most Persistent Pollutants. *Environ. Sci. Technol.* **2016**, *50* (10), 5261−5268.

(275) Margot, J.; Copin, P.-J.; von Gunten, U.; Barry, D. A.; Holliger, C. Sulfamethoxazole and isoproturon degradation and detoxification by a laccase-mediator system: Influence of treatment conditions and mechanistic aspects. *Biochem. Eng. J.* **2015**, *103*, 47−59.

DOI: 10.1021/acs.est.8b00586
Environ. Sci. Technol. 2018, 52, 5062−5075