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## Determination, occurrence, and treatment of saccharin in water: A review

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### Abstract

Saccharin (SAC) is an emerging contaminant, widely detected in the environment, with potential ecotoxicity risks to aqueous organisms and human beings. Wastewater treatment plants (WWTPs) are key sources and sinks of SAC, and play a vital role in eliminating SAC entering the environment. An overview is provided of the potential ecotoxicity of SAC, its occurrence in the aqueous environment, and its degradation performance in WWTPs. SAC treatments, including physical, chemical (mainly advanced oxidation processes AOPs), biological, and hybrid processes, and possible degradation mechanisms are also considered. Of the various SAC removal processes, we find that adsorption-based physical methods exhibit relatively poor performance in terms of SAC removal, whereas chemical methods, especially hydroxy radical-mediated oxidation processes, possess excellent capacities for SAC elimination. Although biological degradation can be efficient at removing SAC, its efficiency depends on oxygen supply and the presence of other co-existing pollutants. Hybrid aerobic biodegradation processes combined with other treatments including AOPs could achieve complete SAC reduction. Furthermore, novel adsorbents, sustainable chemical methods, and bioaugmentation technologies, informed by in-depth studies of degradation mechanisms and the metabolic toxicity of intermediates, are expected further to enhance SAC removal efficiency and enable comprehensive control of SAC potential risks.

**Keywords:** artificial sweeteners; water treatment ;micro-contaminants; absorption; advanced oxidation process; aerobic degradation

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## 1. Background

Artificial sweeteners (ASs) have recently been recognized as emerging micro-contaminants because of their environmental persistence and widespread detection in the aqueous environment (Lange et al., 2012; Scheurer et al., 2010; Weihrauch and Diehl, 2004). ASs are widely consumed in considerable quantities nowadays as sugar substitutes because of their high-intensity sweetness (Kroger et al., 2006; Subedi and Kannan, 2014). Saccharin (SAC) is one of the most popular artificial sweeteners owing to its low price, and sweetness that is about 300 times greater than sugar (Table 1) (Lange et al., 2012). Having been approved by several food and drink regulators, including the Food and Drug Administration, SAC is widely consumed by humans as a calorie-free artificial sweetener in more than 90 countries (Kulkarni, 2018). It is believed that SAC may pass unchanged through the human body, and then reach wastewater treatment plants (WWTPs) as domestic effluent, after which the treated effluent provides the main source of SAC in receiving water bodies from which it can be abstracted and recirculated as drinking/tap water (Buerge et al., 2011; Scheurer et al., 2010). Moreover, SAC and its salts (such as saccharin sodium, also called SAC in the literature) are applied in substantial concentrations in nickel-plating, personal care and pharmaceutical products, antiseptics, preservatives, antistatic agents, feeds, *etc.* (Buerge et al., 2011; Cohen et al., 2008; Rashidi and Amadeh, 2009; Spoelstra et al., 2016; Watanabe et al., 2015; Zygler et al., 2009).

### [Table 1]

SAC has been the focus of a gradually increasing controversy since it was first synthesized in 1879 (Cooper, 1985; Weihrauch and Diehl, 2004). The primary issue associated with SAC was its adverse effects on animals (e.g. carcinogenesis), and therefore probably on humans (Cooper, 1985). Increasingly, environmental and public health concerns have been raised about SAC on account of its long-term application, wide detection in the aqueous environment, and its low, but not negligible, hazard and risk potential to aquatic organisms (Luo et al., 2019). SAC may be a weak carcinogen causing cancer of the urinary tract, even bladder tumors in male rats (Oser, 1985; Uçar and Yılmaz, 2015). SAC-fed zebra fish have experienced elevated levels of cholesteryl ester transfer protein activity in hypercholesterolemia (Kim et al., 2011). SAC may also be one of the main causative factors behind paediatric inflammatory bowel disease, by disturbing gut bacteria (Li et al., 2016; Qin, 2012, 2016, 2019). Previous studies have also suggested that SAC can induce liver inflammation in mice (Bian et al., 2017). Negative effects of SAC have also been observed on plants, where by SAC promotes an increase in the micronucleus of plant roots, the higher concentration indicating greater genotoxicity of certain compounds (Ma et al., 1995) with concomitant cytotoxic and mutagenic impacts on plants (Oliveira et al., 2017). Furthermore, Davididou et al. (2019) found that the degradation products of SAC, having undergone transformations after exposure to solar radiation, were more environmentally toxic than the parent compound SAC, based

on chronic toxicity assessment by monitoring *Vibrio fischeri* bioluminescence. SAC is also a major soil metabolite of certain sulfonylurea herbicides, and so could seep into groundwater, eventually contaminating natural waterbodies such as rivers and lakes (Buerge et al., 2011). Therefore, the elimination of SAC from the aquatic environment is of utmost importance.

A comprehensive study focusing on the fate and mitigation strategies of saccharin in water matrix is still a missing element from literature. Such a study could provide a coherent body of knowledge on the topic, guidance for future research, and guidelines for policymakers and stakeholders. With this in mind, the aim of the present work is to review analytical methods for determining SAC, the occurrence and distribution of SAC in aqueous environment, and the efficiency of several SAC elimination approaches.

## **2. Review methodology**

Given that SAC is hydrophilic, we consider the aqueous environment to act as a key sink of SAC. Taking this perspective, “saccharin”, “water”, and “wastewater” were used as topic words in searching for papers and patents in Web of Science, Scopus, and Google Scholar (as supplementary) without restriction on publication date. Related documents (>100) were selected to summarize research findings on the identification, occurrence, and distribution of SAC in the aqueous environment. We also examined research into applied and potential SAC treatment technologies and their mechanisms for SAC degradation in water and wastewater processes.

## **3. Results and discussion**

### **3.1 Analytical methods for SAC quantification**

A highly-sensitive, accurate method for SAC determination is an important prerequisite for carrying out detailed investigation into the fate of SAC at trace levels from ng/L to µg/L in the aqueous environment. Most recent studies of water and wastewater quality have placed emphasis on physicochemical indicators (such as pH, color, temperature, odor, *etc.*), chemical oxygen demand (COD) content, nitrogen and phosphorus nutrient levels, and concentrations of toxic chemicals (such as heavy metals, persistent organic pollutants (POPs), endocrine disrupting chemicals (EDCs), *etc.*). However, few studies have been reported on the measurement of emerging contaminants with unknown negative effects on environmental and human health (Pal et al., 2014). Given that there is no existing national or international standard method for SAC determination in water samples, a summary of current knowledge on sensitive, accurate methods for SAC determination follows.

To date, analytical techniques for SAC determination in water samples mainly comprise capillary electrophoresis (CE), electroanalysis, flow injection analysis (FIA), gas chromatography (GC), high-performance liquid chromatography (HPLC), reversed-phase HPLC (RP-HPLC), ultra-fast liquid

chromatography (UFLC), ion chromatography (IC), and spectroscopy with thin-layer chromatography (TLC). Different combinations of these analytical techniques have led to more sensitive, accurate methods for determining SAC in water samples or liquids extracted from solid samples. **Table 2** lists the different techniques along with their limits of detection (LODs), and limits of quantitation (LOQs). Determination methods for other ASs or compounds with similar structure to SAC have also been reported and may be considered as alternative SAC determination methods. Examples are LC-ESI-MS/MS (Richter et al., 2007) and ultra HPLC-Q-TOF-MS (UHPLC-Q-TOF-MS)(Russo et al., 2020). Among all the aforementioned techniques, LC-MS/MS has been most widely employed for determining SAC not only in water samples but also in samples prepared by solid phase extraction(SPE) (Arbeláez et al., 2015b; Gan et al., 2013b; Lakade et al., 2018; Ordóñez et al., 2012) or dispersive solid-phase extraction (d-SPE), which are commonly used for extracting target substances from solid phases at concentrations of order mg/L and placing into aqueous samples(Chen et al., 2012; Lakade et al., 2018; Zhao et al., 2013).

**[Table 2]**

For LC-MS/MS, the LODs and LOQs, which indicate instrument sensitivity and accuracy, depend on the performance of the applied liquid chromatography LC and mass spectrometer (MS), the characteristics of the aqueous samples, and operational and environmental factors (**Table 2**). Using SPE-LC-MS/MS, LODs reported for SAC have been in the range of 0.001–0.04 g/L for river water and 0.01–0.5 g/L for influent and effluent wastewater (Arbeláez et al., 2015b). For LC-MS/MS coupled with an electrospray ionization (ESI) source, both the LODs and LOQs were reported to be in the range < 0.015 to 23 ng/L in analyzing wastewater, surface water, and groundwater samples from Vietnam, The Philippines, and Myanmar(Watanabe et al., 2016), and LOD of 0.2 ng/L and LOQ of 0.5 ng/L for surface water and groundwater samples in a separate study(Edwards et al., 2019). For LC-MS/MS using polymeric reversed-phase and mixed-mode with either weak or strong anionic-exchange sorbents, the LOQs for SAC determination obtained for wastewater and surface water samples were in the range 0.01-0.5g/L (Ordóñez et al., 2012). dSPE-UFLC-MS/MS analysis confirmed the presence of nine food additives in red wine, with the results showing that the LOQs for SAC were 0.10-50.0 g/L (Chen et al., 2012). Using HPLC-MS/MS, sensitive detection of SAC in groundwater and surface water was achieved to about 10 ng/L (Berset and Ochsenbein, 2012). However, the LOD of SAC using HPLC-MS/MS after on-line SPE applied to groundwater samples from a landfill site was 0.0003 g/L (Stefania et al., 2019). Gan et al.(2013b) used IP-LC-TQMS to determine artificial sweeteners including SAC, and found that the method quantitation limits varied between 0.4 and 7.5 ng/L for different water samples. Another analytical method for the determination of six artificial sweeteners in sewage sludge, based on pressurised liquid extraction (PLE) with water, followed by SPE and subsequent LC-MS/MS, indicated

that LOQ was 16 ng SAC/g dry weight of sludge (Ordoñez et al., 2013). Kokotou and Thomaidis (2013) used hydrophilic interaction LC-MS/MS to detect and quantify eight artificial sweeteners in wastewater, and reported an instrument LOD of SAC of 0.24 mg/L. Using an isotope-dilution mass spectrometry (IDMS) LC-MS/MS method for quantifying four artificial sweeteners, including SAC, in Finnish surface waters, Perkola and Sainio (2014) recorded LOD of 4.0 ng/L and LOQ of 25.0 ng/L. In an application of LC-MS/MS coupled with hypercross linked particles for extraction of SAC using dSPE from environmental samples, the instrument LODs ranged from 0.05–0.5 µg /L, and the lowest points of the calibration curve were taken as the instrument limits of quantification (ILOQs) (Lakade et al., 2018). Other efficient pre-treatment procedures that have been developed, such as direct-injection (DI) HPLC–MS/MS (Berset et al., 2010; Berset and Ochsenbein, 2012; Ochsenbein et al., 2008), are potential candidates as SAC determination techniques.

### **3.2. Occurrence and distribution of SAC in aqueous environment**

To date, research studies have not only focused on method accuracy and sensitivity but also its applicability to the analysis of actual aqueous matrices containing SAC, such as municipal wastewater, waste activated sludge, surface water, groundwater, and potable water.

**Table 3** summarizes the overall occurrence and distribution of SAC based on current determination methods.

[**Table 3**]

From the information listed in **Table 3** and given in the open literature, it can be seen that WWTPs are a major source of SAC discharged into environmental waters (Subedi et al., 2014). SAC concentrations in the influent of WWTPs vary according to geographical region; for instance, SAC concentrations have been measured as 9.3–389.0 µg/L in Asia, 1.9–25.1 µg/L in North America, and 7.1–18.0 µg/L in Europe (Tran et al., 2018). WWTPs therefore play a vital role in preventing and controlling the ecological environmental risk induced by SAC. Biological treatment processes, especially the conventional activated sludge process and membrane reactor systems, are still the most commonly applied in existing WWTPs. However, although SAC is readily biodegraded, it is still detected in treated effluent from WWTPs and in sewage sludge (waste activated sludge) (Scheurer et al., 2010; Tran et al., 2014b). Consequently, SAC is consistently detected in surface waters receiving effluent from WWTPs, ground water at locations where sewage sludge has been applied as soil fertilizer, and karst aquifers suffering infiltration by wastewater-polluted surface water (Buerge et al., 2011; Edwards et al., 2019). Therefore, there is an urgent need for better technologies to eliminate SACs from the aquatic ecosystem.

### 3.3. Treatment technologies for SAC elimination

To reduce the potential adverse effects and ecotoxicity of SAC on aqueous organisms, efficient approaches are imperative in eliminating SAC from water and wastewater. **Table 4** briefly reviews the present state-of-the-art of SAC treatment technologies, and associated degradation mechanisms, classified in terms of the physical, chemical and biological processes involved. A more detailed description follows.

[Table 4]

#### 3.3.1 Physical methods

Of the physical treatment methods (**Table 4**), adsorption is the most commonly applied process for SAC removal. Adsorption performance relies primarily on adsorbent type(s) and operational conditions, such as pH, and temperature.

##### 3.3.1.1 Activated carbon (AC)

Activated carbon (AC) is an adsorbent that is effective (due to its non-polar nature) at separating many types of chemicals (especially organics) from the water environment, and has been widely utilized in water and wastewater treatment processes (Bernardo et al., 2006). Applications of AC with or without enhancement to SAC elimination have also been investigated (Bernardo et al., 2006; Li et al., 2018b; Mailler et al., 2015; Scheurer et al., 2010). Bernardo et al. (2016) observed that the SAC removal rate by AC adsorption for 16 h increased from 40% to 75% after enhancement by ultrasonication pretreatment at 500 kHz frequency in argon and O<sub>2</sub>/N<sub>2</sub> (20/80 vol%), owing to decomposition of SAC into other compounds that could easily be absorbed by AC when subjected to ultrasound. Li et al. (2018b) achieved a higher removal efficiency of SAC by granular AC (GAC) than for other ASs, such as aspartame (ASP), sucralose (SUC), acesulfame (ACE), cyclamate (CYC), with the removal rate fitting well with SAC's octanol-water partition coefficients -log K<sub>ow</sub> (**Table 1**). Using a filter with powdered AC (PAC), Mailler et al. (2015) achieved SAC removal rates between 33-54%, and observed that efficient removal performance strongly depended on the PAC filter preload, and the combined effects of adsorption by GAC/PAC and biodegradation by microorganisms on GAC/PAC (Scheurer et al., 2010).

##### 3.3.1.2 Bank filtration

Bank filtration is a natural attenuation process that removes or retains SAC in bank soil, based on soil adsorption capacity and simultaneous biodegradation by indigenous microorganisms in the soil. Scheurer et al. (2010) studied the performance of this process in conventional multi-barrier drinking water treatment plants (DWTPs) for ASs removal and found that SAC was eliminated efficiently (nearly 100%) in all waterworks that employed biologically active bank filtration. However, SAC is hydrophilic, and so

biodegradation might contribute more than soil adsorption to the bank filtration process (Buerge et al., 2011). A study of sand (mainly quartz sand and anthracite) filtration in China has also revealed less efficient SAC removal (14.5%) by adsorption of the main filtration media because of the hydrophilic properties of SAC (Gan et al., 2012).

### **3.3.1.3 Resins**

Resins are cost-efficient absorbing materials with porous structures that perform well in ultrapure water preparation and the advanced treatment of micro-contaminants, POPs, EDCs, and heavy metals (Wang et al., 2010). Recently, resins have also been evaluated for SAC removal; Li et al. (2018b) reported that the amount of SAC adsorbed by an applied magnetic anion-exchange resin was 3.33-18.51 times that of GAC under the same experimental conditions. Li et al. also treated three other ASs (ACE, CYC and SUC), and found that resin adsorption was more efficient at ACE removal, illustrating the importance of resin type in the removal of different kinds of ASs.

### **3.3.1.4 Reverse Osmosis (RO)**

Reverse osmosis (RO) is a membrane separation process aimed at the removal of ions and organic matter, and can produce high quality water for reuse (Dialynas et al., 2008; Li et al., 2020). Nevertheless, when RO was tested for SAC removal in an advanced water recycling plant, the observed removal efficiency was unsatisfactory because of the high water solubility of SAC and the proximity of its molecular weight (183 Da) to that of the cut-off molecular weight of the RO membrane (Busetti et al., 2015). Interestingly, RO achieved an excellent removal rate (>97%) for another sweetener, SUC (Lee et al., 2012). This suggests that RO performance for ASs elimination depends on both the molecular weight and the solubility of the ASs. Furthermore, high energy consumption and membrane fouling rates make RO less cost-effective for large-scale water and wastewater treatment applications.

### **3.3.1.5 Other physical approaches**

Novel absorbents, such as highly porous metal-organic framework (MOF) (Seo et al., 2016) and metal azolate framework-6-derived porous carbons (MDCs) (Song et al., 2018) have been used to remove SAC. MOFs modified by urea and melamine and MDCs have achieved high SAC adsorption performances in water treatment processes, in both cases due to H-bonding on the surface of functional groups. Biochar (BC), an absorbent used in decontamination of water and wastewater (Wang and Wang, 2019), was also tested for SAC removal, and relatively higher sorption of SAC was obtained (Liu et al., 2019). It was shown by Inyang and Dickenson that the  $\pi$ - $\pi$  stacking interaction between the SAC aromatic ring and the aromatic structure of graphene in BC made a significant contribution to SAC adsorption. Also, the  $\pi$ - $\pi$  electron donor-acceptor ( $\pi$ - $\pi$  EDA) interaction occurring between electron-withdrawing sulfonamide and carbonyl functional groups ( $\pi$  electron acceptors) in SAC and the  $\pi$ -electron-rich BC



( $\pi$ -electron donors) is also believed to play a part in efficient SAC adsorption by BC (Inyang and Dickenson, 2015). Whereas these results were obtained from bench tests, further studies are needed at pilot-scale and, importantly, full-scale operating conditions. Finally, although other new absorbents (*i.e.*, lignite, xylit) and coagulants (*i.e.*,  $\text{Al}^{3+}$ ) for ASs removal have recently been proposed, their adsorption and/or coagulation capacities for SAC removal have proved relatively low or remain unknown (McKie et al., 2016; Rostvall et al., 2018). This suggests that further selection and application of new absorbents for SAC physical removal should be carried out.

### 3.3.2 Chemical treatments

To date, chemical treatment processes have proved to be the most efficient way to remove organic trace pollutants and thus eliminate their metabolite ecotoxicity. **Table 4** lists the chemical processes used to treat SAC, which are described in more detail below.

#### 3.3.2.1 Chlorination

Chlorination is a widely used disinfection technology applied mainly to inactivate pathogenic microorganisms in DWTPs, water reclamation plants, and WWTPs. (Jia et al., 2015; Li et al., 2017). Studies that introduced chlorination in SAC treatment have found that SAC and other ASs (*i.e.* ACE, CYC, SUC) were persistent and not transformed by the chlorination process, possibly caused by a lack of electron-rich sites for oxidation (Scheurer et al., 2010; Soh et al., 2011; Torres et al., 2011). However, it was found that ACE degradation during chlorination followed pseudo-first-order kinetics and was pH-dependent, and that chlorinated ACE could be the precursor of several regulated disinfection by-products and inhibit *Vibrio fischeri* luminescence (Li et al., 2017). The study also concluded that, although the chlorination process is cost-effective and widely applied, chlorine-related ecotoxic risks should be carefully considered and evaluated. Therefore, more efficient, less eco-risky advanced oxidation processes (AOPs) have been tested for SAC removal; AOPs, including ozonation, UV and Fenton treatment, are based on the *in situ* generation of strong oxidants, mainly hydroxyl-radicals ( $\cdot\text{OH}$ ), for the oxidation of organic compounds (Miklos et al., 2018).

#### 3.3.2.2 Ozonation

Ozonation is one of the most robust advanced oxidation processes (AOPs) for eliminating organic contaminants in wastewater (Xiao et al., 2015). The primary oxidation mechanisms for ozonation are pH-dependent: in acidic conditions, ozone acts as the main oxidant and can react directly with organics, whereas in neutral and alkaline conditions, radicals (mainly  $\cdot\text{OH}$ ) are formed, initiating oxidation reactions (Pak et al., 2016). Ozone can oxidize a wide range of organics, including those containing aromatic and/or amine-groups (Comninellis et al., 2008; Hollender et al., 2009; Stefan, 2017), and so it has been adopted to remove ASs. A study of multi-barrier DWTPs reported low performance for SAC

removal when reacting directly with ozone instead of its radicals, whereas ozonation was found to degrade ACE well, with moderate removal of SUC, depending on the operating conditions, including ozone concentration and co-existing competing contaminants (Scheurer et al., 2010; Shao et al., 2019). Similar test results indicated that SUC could be degraded significantly through ozonation by radical-mediated oxidation with a relatively high ozone dose (Hu et al., 2017; Lee et al., 2012; Torres et al., 2011). The higher removal efficiencies of ACE and SUC than of SAC might be attributed to the higher oxidation capacity of  $\cdot\text{OH}$  ( $\sim 2.80$  V) compared with ozone ( $\sim 2.07$  V).

### 3.3.2.3 Ultraviolet (UV) irradiation

Ultraviolet (UV) irradiation has been applied to the disinfection and photolysis of organics, exploiting the fact that radiation energy in the ultraviolet (UV) region of the electromagnetic spectrum can alter the chemical bonds of organics (Sastry et al., 2000). Although UV irradiation has recently been tested for SAC removal, it was found invalid because of the low molar extinction coefficient of SAC (Buseti et al., 2015; Li et al., 2018b; Perkola et al., 2016). Even so, photocatalytic oxidation, (*i.e.* UV irradiation combined with other oxidants and/or catalysts) has proved efficient for SAC elimination because additional  $\cdot\text{OH}$  ions are generated which in turn oxidize organics in water. Li et al. (2018b) compared ASs oxidation processes involving UV/H<sub>2</sub>O<sub>2</sub> and UV/sodium peroxydisulfate (PDS) in five full-scale WWTPs for removal of four ASs, including SAC; the influent SAC concentrations ranged from 430 to 27,340 ng/L, and the degradation efficiency was similar for all ASs. However, it has been reported that UV/PDS performed better than UV/H<sub>2</sub>O<sub>2</sub> with the same oxidant dosage because SO<sub>4</sub><sup>•-</sup> generated from UV/PDS has stronger redox potential (up to 3.10 V at neutral pH, and  $\cdot\text{OH}$  of 2.80 V from UV/H<sub>2</sub>O<sub>2</sub>) and higher selectivity to organic compounds (Xu et al., 2016). Davididou et al. (2017) investigated photocatalytic treatment of SAC under UV irradiation in the presence of TiO<sub>2</sub> nanocomposites tuned by Sn (IV) (which absorbs UV light to produce electrons near the conduction band and holes near the valence band (El-Mekkawi et al., 2020)), and found that photocatalytic performance correlated positively with catalyst loading (up to 400 mg/L) for SAC removal, following a pseudo-first order kinetic rate. Davididou et al. (2018) also assessed the performances of UV-light-emitting diode (LED) and conventional UV-black light (BL) technologies for SAC removal in the presence of TiO<sub>2</sub>, exploiting the generation of highly reactive species (mainly  $\cdot\text{OH}$ ) when photocatalysis is initiated by UV illumination on TiO<sub>2</sub>. Data analysis showed that UV-LED was 16 times more energy efficient than UV-BL (Davididou et al., 2018); the enhanced performance of UV-LED was most likely due to the addition of hydroxyl radicals in the aromatic ring, leading to hydroxylated products during photocatalytic transformation (Toth et al., 2012). A contemporaneous study (Zelinski et al., 2018) observed that more than 99% of SAC could be degraded and up to 49% mineralized by TiO<sub>2</sub>/UV-A, and, importantly, demonstrated the low toxicity of the photocatalyzed solution of SAC using the *Artemiasalina* test. Zelinski et al. also found that UV-LED

in the presence of  $\text{TiO}_2$  also achieved very high degradation efficiency (up to 90%) of ACE compared with other photocatalytic processes using UV-LED with  $\text{ZnO}$ ,  $\text{H}_2\text{O}_2$ , peroxomonosulfate (PMS) and PDS, respectively. Furthermore, the ACE degradation rate in UV-LED photolysis process could be enhanced by addition of catalysts and/or oxidants (Wang et al., 2019). Further pilot tests are clearly needed concerning the cost-effectiveness of UV-LED and water ecotoxicity during the oxidation process.

#### **3.3.2.4 Fenton treatment**

The Fenton reaction, a widely applied AOP for removal of organics and inorganics from the aqueous environment, was discovered by H.J.H. Fenton in 1894 and consists of reactions of peroxides (mainly  $\text{H}_2\text{O}_2$ ) with ferrous ions ( $\text{Fe}^{2+}$ ) to generate active species (usually  $\cdot\text{OH}$ ) as strong oxidants for the oxidation degradation of pollutants (Fenton, 1894). To date, many attempts have been made to treat wastewater using the Fenton (or Fenton-like) reaction, whereby ferric ions ( $\text{Fe}^{3+}$ ) react with  $\text{H}_2\text{O}_2$  to produce  $\text{Fe}^{2+}$ , thus promoting oxidation (Goldstein et al., 1993; Kahoush et al., 2018; Wang, 2008). Recently, the Fenton (or Fenton-like) reaction has been applied to SAC degradation. Chen et al. (2014) utilized photo-Fenton oxidation to degrade saccharin sodium, and reported that TOC decreased by 93% under optimal conditions (500W mercury lamp, 0.04 g  $\text{TiO}_2$ , pH=7, and photo-Fenton ratio of SAC: $\text{H}_2\text{O}_2$ : $\text{Fe}^{2+}$ =5:60:1). Davididou et al. (2019) investigated SAC removal in a solar compound parabolic collector pilot plant using conventional photo-Fenton at pH 2.8 and olive mill wastewater (OMW)-assisted photo-Fenton. Although higher SAC degradation was achieved by conventional photo-Fenton, encouraging results were obtained using the more sustainable photo-Fenton process with OMW as an iron chelating agent. Moreover, Lin et al. (2016) employed an electro-Fenton process to remove SAC from the aqueous environment, and observed that the SAC degradation rate driven by electrochemically generated  $\cdot\text{OH}$  ions followed pseudo-first order kinetics in the presence of Pt and a boron-doped diamond anode. Importantly, the toxicity of treated solution with oxalic, formic, and maleic acid as aliphatic byproducts of SAC was also eliminated after 60-min electro-Fenton reaction (Lin et al., 2016). Excellent ASP degradation performance can be achieved using electro-Fenton reaction, depending on  $\text{Fe}^{2+}$  concentration and applied current (Richards et al., 2017). Nowadays, growing attention is being paid to sustainable Fenton processes, such as Bio-Fenton, Bio-Electro-Fenton, and Electrochemical/ $\alpha$ -FeOOH/PDS approaches; however, further investigations are required in order to better evaluate their SAC degradation efficiencies (Kahoush et al., 2018; Li et al., 2019).

All in all, although AOPs appear attractive as effective technologies for SAC elimination, their performance should be assessed not only in terms of SAC removal, but also in terms of ecotoxicity variation during reaction processes.

#### **3.3.3. Biological degradation and uptake**

Biodegradation and uptake, comprising degradation, assimilation, and utilization during biological

metabolisms or activities, are cost-effective, eco-friendly processes that are widely applied for contaminants removal from environmental media (Gaur et al., 2018). Several studies, listed in **Table 4**, have investigated the SAC biodegradable potential of pure strains, mixed microbial communities, and environmental media with rich biodiversity (Richards et al., 2017; Scheurer et al., 2014; Slavič et al., 2018).

### **3.3.3.1 Bacteria and plants**

As reported in 2003, anaerobic bacterium strain, *Spingomonas xenophaga* SKN, was isolated from domestic sewage and proved to degrade SAC well, with catechol providing the sole source of carbon and energy for growth (Schleheck and Cook, 2003). Later, it was discovered that efficient degradation of SAC involved the participation of both heterotrophic and autotrophic nitrifying microorganisms along with various non-specific oxidative enzymes, and that there was an observable linear relationship between nitrification rate and co-metabolic biodegradation rate (Tran et al., 2014b). *Coliform* bacteria and spoilage bacteria have been identified as being responsible for SAC degradation in drinks (Kabir et al., 2018). Besides, there is a growing body of research suggesting that a substantial proportion of SAC could be degraded by soil microorganisms (Biel-Maeso et al., 2019; Richards et al., 2017; Van Stempvoort et al., 2011a). Recent genomic resolution of bacterial populations in SAC degradation processes has revealed that the dominant saccharin-degrading consortia comprised  $\beta$ -proteobacterial genome from the *Rhodocyclaceae* family (Deng et al., 2019). Larger-scale plants have also proved effective at SAC removal from contaminated soil (Baalbaki et al., 2017). The foregoing indicates an opportunity whereby microorganisms and plants could be isolated and/or enriched in water and applied in wastewater treatment processes to efficient degradation of SAC pollutants.

### **3.3.3.2 Activated sludge/consortia**

The behavior of SAC in water and wastewater treatment processes, especially biological processes encountered in activated sludge systems and membrane bioreactors, has been studied extensively. Buerge et al. (2009) provided evidence for the high aerobic biodegradation of SAC (mean elimination efficiency of 90%, confirmed by Subedi and Kannan (2014) and Tran et al. (2015)) in WWTP activated sludge incubation experiments, with no elimination observed in sterile conditions. Buerge et al. also found that ACE and SUC exhibited persistence with no degradation within 7h, whereas SAC was degraded by 78% in 3h under the same operating conditions. Another systematic, bench-scale study (Scheurer et al., 2010) found SAC to be more readily biodegradable in a fixed-bed bioreactor, with complete elimination within 20 d, but hardly any degradation of ACE and SUC even after 92 d. Research on SAC removal in artificial wetlands has also provided evidence that SAC removal is positively correlated ( $p < 0.05$ ) to dissolved oxygen concentration (Vymazal and Dvořáková Březinová, 2016). Complete SAC removal has also been observed in sequencing batch reactors (SBRs), where the presence of SAC had no impact on the removals

of COD, ammonia nitrogen, and total nitrogen (Li et al., 2018a). In a study of primary sludge anaerobic digestion, Phan et al. (2018) observed that SAC was mainly removed in the aqueous phase during anaerobic digestion. In a similar study, Buerge et al. (2011) measured more than 95% SAC removal during the fermentation process, but no SAC removal from the digested sewage sludge. Differences between the microbial communities in the two foregoing studies might be the main reason for the dissimilar SAC removal efficiencies. Future research is needed assessing factors influencing SAC biodegradation performance (e.g. temperature, pH, and hydrodynamics) in treatment units for saccharin sodium and other ASs (Baalbaki et al., 2017; Qu et al., 2019; Shreve and Brennan, 2019).

Overall, biodegradation plays an important part in aqueous SAC treatment, and more advanced molecular biotechnologies could be used to improve efficiency of biological SAC removal. However, given that SAC can accumulate in sludge (Subedi et al., 2014), more attention should be paid to sludge treatment *per se*, which is another important environmental issue worldwide.

### **3.4. Integrated processes**

As described above, treatment approaches based on physical, chemical, and biological principles have particular merits and drawbacks for SAC removal. Hybrid processes, also known as multi-technology processes, are being applied to eliminate SAC ecotoxicity in DWTPs and WWTPs. A popular combination of processes comprises flocculation, sand filtration, AC, or biological treatment (e.g. biosorption, biodegradation), and disinfection by chlorination, ozonation, or UV irradiation in DWTPs (Gan et al., 2012; McKie et al., 2016; Scheurer et al., 2010). Primary (flocculation, sedimentation, *etc.*), secondary (mainly biological degradation under aerobic, anoxic, or anaerobic conditions), and tertiary (disinfection using chlorine, ozone and UV) treatments are the three serial processes that constitute many WWTPs (Buerge et al., 2009; Li et al., 2018b; Subedi and Kannan, 2014; Tran et al., 2015).

For DWTPs, although SAC removal through adsorption is not efficient, the biodegradation of SAC performs well during filtration with active microorganisms located in and on the surface of porous filters (Buerge et al., 2011; Gan et al., 2012; Scheurer et al., 2010). The foregoing studies reported that disinfection by chlorination, ozonation, and UV irradiation exhibited poor/moderate SAC removal, but achieved better degradation performance of ACE.

For WWTPs, more than 90% of SAC could be removed in primary and secondary processes, with biodegradation a key mechanism; however SAC in tertiary treatment would undergo less transformation than other ASs, such as ACE and SUC (Buerge et al., 2009; Li et al., 2018b; Subedi and Kannan, 2014; Tran et al., 2015). This suggests that improving SAC biodegradation efficiency is of great importance in water and wastewater treatment practice.

### **3.5 Future research**

Following this extended literature review on SAC detection, occurrence, and treatment in water,

recommendations for future studies can be drawn, as described below. Given that WWTPs are a major source of SAC discharged into environmental waters (Subedi et al., 2014), emphasis should be placed on developing an efficient technology to eliminate trace amounts of SAC. Advanced oxidation and biological degradation processes seem presently to be a better option than physical processes because of the high hydrophilicity of SACs. During advanced oxidation it is important to evaluate the eco-toxicity of intermediate metabolites from SAC degradation along with its removal efficiency, noting that toxicity is linked with the presence of degradation products (Davididou et al., 2019). Also, further studies are needed to investigate the interaction relationship between SAC and co-existing pollutants, given that SAC co-exists in practice with other pollutants, such as nutrients, ACE, SUC, and other emerging pollutants, some of which exhibit strong reactivity (Scheurer et al., 2010; Shao et al., 2019; Tran et al., 2014b). Inspired by the synchronous biodegradation of various pollutants in soil and activated sludge, biological processes offer a great opportunity in practice as cost-effective SAC removal methods; further research is recommended to isolate certain functional strains to enrich dominant biological communities that are key to water and wastewater treatment processes (Baalbaki et al., 2017; Biel-Maeso et al., 2019).

#### **4. Conclusion**

This state-of-the-art review has examined the current understanding of SAC degradation processes and the occurrence and distribution of SAC, determined primarily by LC-MS/MS and other novel or modified technologies. As an emerging contaminant, SAC appears not as safe as believed previously, owing to its worldwide long-term, large volume consumption, its ubiquitous detection in environmental media, and its potential chronic eco-toxicity. It was observed that although there is no existing national or international standard method for SAC determination in water samples, accurate methods for SAC determination have been recently developed. Based on these analytical techniques, several studies confirmed the occurrence of SAC at WWTP discharges worldwide at the  $\mu\text{g/L}$  order, indicating that treatment plants play a vital role in preventing and controlling the ecological environmental risk induced by SAC. Extensive research on physical, chemical, and biological technologies for SAC has been carried out. Among these techniques, advanced oxidation and biological processes appear to be efficient ways to eliminate SAC. Regarding future research, the focus should be placed on the ecotoxicity of SAC degradation products as well as its interaction with other co-existing water pollutants. Moreover, the isolation and application of functional strains to enrich dominant biological communities may be key to development of successful biological processes for wastewater treatment. In all cases, a cost-benefit analysis of SAC treatment would be recommended before scaling up the process.

#### **Declaration of competing interest**

The authors declare no conflict of interest.

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## References

- Arbeláez, P., Borrull, F., Maria Marcé, R., Pocurull, E., 2015a. Trace-level determination of sweeteners in sewage sludge using selective pressurized liquid extraction and liquid chromatography–tandem mass spectrometry. *Journal of Chromatography A* 1408, 15-21. <https://doi.org/10.1016/j.chroma.2015.07.001>
- Arbeláez, P., Borrull, F., Pocurull, E., Marcé, R.M., 2015b. Determination of high-intensity sweeteners in river water and wastewater by solid-phase extraction and liquid chromatography–tandem mass spectrometry. *Journal of Chromatography A* 1393, 106-114. <https://doi.org/10.1016/j.chroma.2015.03.035>
- Baalbaki, Z., Sultana, T., Metcalfe, C., Yargeau, V., 2017. Estimating removals of contaminants of emerging concern from wastewater treatment plants: The critical role of wastewater hydrodynamics. *Chemosphere* 178, 439-448. <https://doi.org/10.1016/j.chemosphere.2017.03.070>
- Bergamo, A.B., Fracassi da Silva, J.A., de Jesus, D.P., 2011. Simultaneous determination of aspartame, cyclamate, saccharin and acesulfame-K in soft drinks and tabletop sweetener formulations by capillary electrophoresis with capacitively coupled contactless conductivity detection. *Food Chemistry* 124(4), 1714-1717. <https://doi.org/10.1016/j.foodchem.2010.07.107>
- Bernardo, E.C., Fukuta, T., Fujita, T., Ona, E.P., Kojima, Y., Matsuda, H., 2006. Enhancement of saccharin removal from aqueous solution by activated carbon adsorption with ultrasonic treatment. *Ultrasonics Sonochemistry* 13(1), 13-18. <https://doi.org/10.1016/j.ultsonch.2004.12.007>
- Berset, J.-D., Brenneisen, R., Mathieu, C., 2010. Analysis of llicit and illicit drugs in waste, surface and lake water samples using large volume direct injection high performance liquid chromatography – Electro spray tandem mass spectrometry (HPLC–MS/MS). *Chemosphere* 81(7), 859-866. <https://doi.org/10.1016/j.chemosphere.2010.08.011>
- Berset, J.-D., Ochsenbein, N., 2012. Stability considerations of aspartame in the direct analysis of artificial sweeteners in water samples using high-performance liquid chromatography–tandem mass spectrometry (HPLC–MS/MS). *Chemosphere* 88(5), 563-569. <https://doi.org/10.1016/j.chemosphere.2012.03.030>
- Bian, X., Tu, P., Chi, L., Gao, B., Ru, H., Lu, K., 2017. Saccharin induced liver inflammation in mice by altering the gut microbiota and its metabolic functions. *Food and Chemical Toxicology* 107, 530-539. <https://doi.org/10.1016/j.fct.2017.04.045>
- Biel-Maeso, M., González-González, C., Lara-Martín, P.A., Corada-Fernández, C., 2019. Sorption and degradation of contaminants of emerging concern in soils under aerobic and anaerobic conditions. *Science of The Total Environment* 666, 662-671. <https://doi.org/10.1016/j.scitotenv.2019.02.279>
- Brumovský, M., Bečanová, J., Kohoutek, J., Borghini, M., Nizzetto, L., 2017. Contaminants of emerging concern in the open sea waters of the Western Mediterranean. *Environmental Pollution* 229, 976-983. <https://doi.org/10.1016/j.envpol.2017.07.082>
- Buerge, I.J., Buser, H.-R., Kahle, M., Müller, M.D., Poiger, T., 2009. Ubiquitous occurrence of the artificial sweetener acesulfame in the aquatic environment: an ideal chemical marker of domestic wastewater in groundwater. *Environmental Science & Technology* 43(12), 4381-4385. [10.1021/es900126x](https://doi.org/10.1021/es900126x)
- Buerge, I.J., Keller, M., Buser, H.-R., Müller, M.D., Poiger, T., 2011. Saccharin and other artificial sweeteners in soils: estimated inputs from agriculture and households, degradation, and leaching to groundwater. *Environmental Science & Technology* 45(2), 615-621. [10.1021/es1031272](https://doi.org/10.1021/es1031272)
- Busetti, F., Ruff, M., Linge, K.L., 2015. Target screening of chemicals of concern in recycled water. *Environ Sci-Wat Res* 1(5), 659-667. <https://doi.org/10.1039/C4EW00104D>



Chen, S.W., Li, W.C., Sun, Z.G., Xie, H.Y., 2014. Degradation of artificial sweetener saccharin sodium by advanced oxidation technology. *Applied Mechanics and Materials* 448-453, 7-10.10.4028/[www.scientific.net/AMM.448-453.7](http://www.scientific.net/AMM.448-453.7)

Chen, X.-H., Zhao, Y.-G., Shen, H.-Y., Jin, M.-C., 2012. Application of dispersive solid-phase extraction and ultra-fast liquid chromatography–tandem quadrupole mass spectrometry in food additive residue analysis of red wine. *Journal of Chromatography A* 1263, 34-42.<https://doi.org/10.1016/j.chroma.2012.09.074>

Cohen, S.M., Arnold, L.L., Emerson, J.L., 2008. Safety of saccharin. *Agro Food Industry Hi-Tech* 19, 26-29

Comninellis, C., Kapalka, A., Malato, S., Parsons, S.A., Poullos, I., Mantzavinos, D., 2008. Advanced oxidation processes for water treatment: advances and trends for R&D. *Journal of Chemical Technology & Biotechnology* 83(6), 769-776.10.1002/jctb.1873

Cooper, R.M., 1985. Saccharin-Of Risk and Democracy. *Food, Drug, Cosmetic Law Journal* 40, 34

Davididou, K., Chatzisyneon, E., Perez-Estrada, L., Oller, I., Malato, S., 2019. Photo-Fenton treatment of saccharin in a solar pilot compound parabolic collector: Use of olive mill wastewater as iron chelating agent, preliminary results. *Journal of Hazardous Materials* 372, 137-144.<https://doi.org/10.1016/j.jhazmat.2018.03.016>

Davididou, K., Hale, E., Lane, N., Chatzisyneon, E., Pichavant, A., Hochepped, J.F., 2017. Photocatalytic treatment of saccharin and bisphenol-A in the presence of TiO<sub>2</sub> nanocomposites tuned by Sn(IV). *Catalysis Today* 287, 3-9.<https://doi.org/10.1016/j.cattod.2017.01.038>

Davididou, K., McRitchie, C., Antonopoulou, M., Konstantinou, I., Chatzisyneon, E., 2018. Photocatalytic degradation of saccharin under UV-LED and blacklight irradiation. *Journal of Chemical Technology & Biotechnology* 93(1), 269-276.10.1002/jctb.5349

Deng, Y., Wang, Y., Xia, Y., Zhang, A.N., Zhao, Y., Zhang, T., 2019. Genomic resolution of bacterial populations in saccharin and cyclamate degradation. *Science of The Total Environment* 658, 357-366.<https://doi.org/10.1016/j.scitotenv.2018.12.162>

Dialynas, E., Mantzavinos, D., Diamadopoulou, E., 2008. Advanced treatment of the reverse osmosis concentrate produced during reclamation of municipal wastewater. *Water Research* 42(18), 4603-4608.<https://doi.org/10.1016/j.watres.2008.08.008>

Edwards, Q.A., Kulikov, S.M., Garner-O'Neale, L.D., Metcalfe, C.D., Sultana, T., 2017. Contaminants of emerging concern in surface waters in Barbados, West Indies. *Environmental Monitoring and Assessment* 189(12), 636.10.1007/s10661-017-6341-4

Edwards, Q.A., Sultana, T., Kulikov, S.M., Garner-O'Neale, L.D., Metcalfe, C.D., 2019. Micropollutants related to human activity in groundwater resources in Barbados, West Indies. *Science of The Total Environment* 671, 76-82.<https://doi.org/10.1016/j.scitotenv.2019.03.314>

El-Mekkawi, D.M., Abdelwahab, N.A., Mohamed, W.A.A., Taha, N.A., Abdel-Mottaleb, M.S.A., 2020. Solar photocatalytic treatment of industrial wastewater utilizing recycled polymeric disposals as TiO<sub>2</sub> supports. *Journal of Cleaner Production* 249, 119430.<https://doi.org/10.1016/j.jclepro.2019.119430>

Fatibello-Filho, O., Aniceto, C., 1999. Flow injection potentiometric determination of saccharin in dietary products using a tubular ion-selective electrode. *Laboratory Robotics and Automation* 11(4), 234-239.10.1002/(SICI)1098-2728(1999)11:4<234::AID-LRA9>3.0.CO;2-A

Fatibello-Filho, O., Nóbrega, J.A., Moraes Guaritá-Santos, A.J., 1994. Flow injection potentiometric determination of saccharin in dietary products with relocation of filtration unit. *Talanta* 41(5), 731-734.[https://doi.org/10.1016/0039-9140\(93\)E0018-9](https://doi.org/10.1016/0039-9140(93)E0018-9)

- Fenton, H., 1894. Oxidation of tartaric acid in presence of iron. *Journal of the Chemical Society, Transactions* 65, 899-910
- Ferrer, I., Thurman, E.M., 2010. Analysis of sucralose and other sweeteners in water and beverage samples by liquid chromatography/time-of-flight mass spectrometry. *Journal of Chromatography A* 1217(25), 4127-4134. <https://doi.org/10.1016/j.chroma.2010.02.020>
- Frazier, R.A., Inns, E.L., Dossi, N., Ames, J.M., Nursten, H.E., 2000. Development of a capillary electrophoresis method for the simultaneous analysis of artificial sweeteners, preservatives and colours in soft drinks. *Journal of Chromatography A* 876(1), 213-220. [https://doi.org/10.1016/S0021-9673\(00\)00184-9](https://doi.org/10.1016/S0021-9673(00)00184-9)
- Gan, Z., Sun, H., Feng, B., 2012. Fate of artificial sweeteners in waste water and drinking water treatment processes (in Chinese). *Research of Environmental Sciences* 25(11), 1250-1256. [10.13198/j.res.2012.11.64.ganzhw.002](https://doi.org/10.13198/j.res.2012.11.64.ganzhw.002)
- Gan, Z., Sun, H., Feng, B., Wang, R., Zhang, Y., 2013a. Occurrence of seven artificial sweeteners in the aquatic environment and precipitation of Tianjin, China. *Water Research* 47(14), 4928-4937. <https://doi.org/10.1016/j.watres.2013.05.038>
- Gan, Z., Sun, H., Wang, R., Feng, B., 2013b. A novel solid-phase extraction for the concentration of sweeteners in water and analysis by ion-pair liquid chromatography–triple quadrupole mass spectrometry. *Journal of Chromatography A* 1274, 87-96. <https://doi.org/10.1016/j.chroma.2012.11.081>
- Gan, Z., Sun, H., Yao, Y., Zhao, Y., Li, Y., Zhang, Y., Hu, H., Wang, R., 2014. Distribution of artificial sweeteners in dust and soil in China and their seasonal variations in the environment of Tianjin. *Science of The Total Environment* 488-489, 168-175. <https://doi.org/10.1016/j.scitotenv.2014.04.084>
- Gaur, N., Narasimhulu, K., Y, P., 2018. Recent advances in the bio-remediation of persistent organic pollutants and its effect on environment. *Journal of Cleaner Production* 198, 1602-1631. <https://doi.org/10.1016/j.jclepro.2018.07.076>
- Goldstein, S., Meyerstein, D., Czapski, G., 1993. The Fenton reagents. *Free Radical Biology and Medicine* 15(4), 435-445. [https://doi.org/10.1016/0891-5849\(93\)90043-T](https://doi.org/10.1016/0891-5849(93)90043-T)
- Hernández, F., Ibáñez, M., Botero-Coy, A.-M., Bade, R., Bustos-López, M.C., Rincón, J., Moncayo, A., Bijlsma, L., 2015. LC-QTOF MS screening of more than 1,000 licit and illicit drugs and their metabolites in wastewater and surface waters from the area of Bogotá, Colombia. *Analytical and Bioanalytical Chemistry* 407(21), 6405-6416. [10.1007/s00216-015-8796-x](https://doi.org/10.1007/s00216-015-8796-x)
- Holder, C.L., Bowman, M.C., 1980. Determination of sodium saccharin in animal feed, wastewater and human urine by high-pressure liquid chromatography. *Toxicology Letters* 5(1), 27-38. [https://doi.org/10.1016/0378-4274\(80\)90144-7](https://doi.org/10.1016/0378-4274(80)90144-7)
- Hollender, J., Zimmermann, S.G., Koepke, S., Krauss, M., McArdell, C.S., Ort, C., Singer, H., von Gunten, U., Siegrist, H., 2009. Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. *Environmental Science & Technology* 43(20), 7862-7869. [10.1021/es9014629](https://doi.org/10.1021/es9014629)
- Hu, R., Zhang, L., Hu, J., 2017. Investigation of ozonation kinetics and transformation products of sucralose. *Science of The Total Environment* 603-604, 8-17. <https://doi.org/10.1016/j.scitotenv.2017.06.033>
- Inyang, M., Dickenson, E., 2015. The potential role of biochar in the removal of organic and microbial contaminants from potable and reuse water: A review. *Chemosphere* 134, 232-240. <https://doi.org/10.1016/j.chemosphere.2015.03.072>
- Jia, S., Shi, P., Hu, Q., Li, B., Zhang, T., Zhang, X.-X., 2015. Bacterial community shift drives antibiotic

resistance promotion during drinking water chlorination. *Environmental Science & Technology* 49(20), 12271-12279.10.1021/acs.est.5b03521

Kabir, B.A., Farouq, A., Ibrahim, A., Rabi'u, A., Bala, A., Mumuney, K., Salisu, H., Abdullahi, S., 2018. Evaluation of the Fate of Saccharin during Storage of Sobo Drink. *Journal of Advances in Microbiology*, 1-8.<https://doi.org/10.9734/JAMB/2018/45704>

Kahoush, M., Behary, N., Cayla, A., Nierstrasz, V., 2018. Bio-Fenton and Bio-electro-Fenton as sustainable methods for degrading organic pollutants in wastewater. *Process Biochemistry* 64, 237-247.<https://doi.org/10.1016/j.procbio.2017.10.003>

Kempińska, D., Kot-Wasik, A., 2018. The use of RP-HPLC–Q-TOF–MS as a powerful tool for wastewater composition profiling and selection of water pollution marker specific to wastewater contamination. *Monatshefte für Chemie - Chemical Monthly* 149(9), 1595-1604.10.1007/s00706-018-2259-y

Kim, J.-Y., Seo, J., Cho, K.-H., 2011. Aspartame-fed zebrafish exhibit acute deaths with swimming defects and saccharin-fed zebrafish have elevation of cholesteryl ester transfer protein activity in hypercholesterolemia. *Food and Chemical Toxicology* 49(11), 2899-2905.<https://doi.org/10.1016/j.fct.2011.08.001>

Kokotou, M.G., Thomaidis, N.S., 2013. Determination of eight artificial sweeteners in wastewater by hydrophilic interaction liquid chromatography-tandem mass spectrometry. *Analytical Methods* 5(16), 3825-3833.10.1039/C3AY40599K

Kroger, M., Meister, K., Kava, R., 2006. Low-calorie sweeteners and other sugar substitutes: a review of the safety issues. *Comprehensive Reviews in Food Science and Food Safety* 5(2), 35-47.10.1111/j.1541-4337.2006.tb00081.x

Kulkarni, C., 2018. Determination of the saccharin content in some ice creams available in market. *Int. J. Food Sci. Nutr* 3(1), 158-160

Lakade, S.S., Zhou, Q., Li, A., Borrull, F., Fontanals, N., Marcé, R.M., 2018. Hypercrosslinked particles for the extraction of sweeteners using dispersive solid-phase extraction from environmental samples. *Journal of Separation Science* 41(7), 1618-1624.10.1002/jssc.201701113

Lange, F.T., Scheurer, M., Brauch, H., 2012. Artificial sweeteners—a recently recognized class of emerging environmental contaminants: a review. *Analytical bioanalytical chemistry* 403(9), 2503-2518

Lee, C.O., Howe, K.J., Thomson, B.M., 2012. Ozone and biofiltration as an alternative to reverse osmosis for removing PPCPs and micropollutants from treated wastewater. *Water Research* 46(4), 1005-1014.<https://doi.org/10.1016/j.watres.2011.11.069>

Li, A.J., Wu, P., Law, J.C.-F., Chow, C.-H., Postigo, C., Guo, Y., Leung, K.S.-Y., 2017. Transformation of acesulfame in chlorination: Kinetics study, identification of byproducts, and toxicity assessment. *Water Research* 117, 157-166.<https://doi.org/10.1016/j.watres.2017.03.053>

Li, J., Li, Y., Xiong, Z., Yao, G., Lai, B., 2019. The electrochemical advanced oxidation processes coupling of oxidants for organic pollutants degradation: A mini-review. *Chinese Chemical Letters* 30(12), 2139-2146.<https://doi.org/10.1016/j.ccllet.2019.04.057>

Li, R., Zheng, J., Jiang, M., Liu, Y., Qin, X., Wang, X., 2016. Increased digestive proteases and decreased  $\beta$ -glucuronidase in feces of rats treated with sucralose and saccharin—another critical evidence that these dietary chemicals may be important causative factors for inflammatory bowel disease. *Inflammatory bowel diseases* 22(8), E29-E30.<https://doi.org/10.1097/MIB.0000000000000859>

Li, S., Geng, J., Wu, G., Gao, X., Fu, Y., Ren, H., 2018a. Removal of artificial sweeteners and their effects on microbial communities in sequencing batch reactors. *Scientific Reports* 8(1), 3399.10.1038/s41598-

018-21564-x

Li, S., Ren, Y., Fu, Y., Gao, X., Jiang, C., Wu, G., Ren, H., Geng, J., 2018b. Fate of artificial sweeteners through wastewater treatment plants and water treatment processes. *PloS one* 13(1), e0189867.<https://doi.org/10.1371/journal.pone.0189867>

Li, Y., Li, M., Xiao, K., Huang, X., 2020. Reverse osmosis membrane autopsy in coal chemical wastewater treatment: Evidences of spatially heterogeneous fouling and organic-inorganic synergistic effect. *Journal of Cleaner Production* 246, 118964.<https://doi.org/10.1016/j.jclepro.2019.118964>

Lin, H., Wu, J., Oturan, N., Zhang, H., Oturan, M.A., 2016. Degradation of artificial sweetener saccharin in aqueous medium by electrochemically generated hydroxyl radicals. *Environmental Science and Pollution Research* 23(5), 4442-4453.[10.1007/s11356-015-5633-x](https://doi.org/10.1007/s11356-015-5633-x)

Liu, Y., Blowes, D.W., Ptacek, C.J., Groza, L.G., 2019. Removal of pharmaceutical compounds, artificial sweeteners, and perfluoroalkyl substances from water using a passive treatment system containing zero-valent iron and biochar. *Science of The Total Environment* 691, 165-177.<https://doi.org/10.1016/j.scitotenv.2019.06.450>

Luo, J., Zhang, Q., Cao, M., Wu, L., Cao, J., Fang, F., Li, C., Xue, Z., Feng, Q., 2019. Ecotoxicity and environmental fates of newly recognized contaminants-artificial sweeteners: A review. *Science of The Total Environment* 653, 1149-1160.<https://doi.org/10.1016/j.scitotenv.2018.10.445>

Ma, T.-H., Xu, Z., Xu, C., McConnell, H., Valtierra Rabago, E., Adriana Arreola, G., Zhang, H., 1995. The improved Allium/Vicia root tip micronucleus assay for clastogenicity of environmental pollutants. *Mutation Research/Environmental Mutagenesis and Related Subjects* 334(2), 185-195.[https://doi.org/10.1016/0165-1161\(95\)90010-1](https://doi.org/10.1016/0165-1161(95)90010-1)

Mailler, R., Gasperi, J., Coquet, Y., Deshayes, S., Zedek, S., Cren-Olivé, C., Cartiser, N., Eudes, V., Bressy, A., Caupos, E., Moilleron, R., Chebbo, G., Rocher, V., 2015. Study of a large scale powdered activated carbon pilot: Removals of a wide range of emerging and priority micropollutants from wastewater treatment plant effluents. *Water Research* 72, 315-330.<https://doi.org/10.1016/j.watres.2014.10.047>

Mali, B., Rathod, D., Garad, M., 2005. Thin-layer chromatographic determination of diazepam, phenobarbitone, and saccharin in toddy samples. *JPC-Journal of Planar Chromatography-Modern TLC* 18(104), 330-332.<https://doi.org/10.1556/JPC.18.2005.4.16>

McKie, M.J., Andrews, S.A., Andrews, R.C., 2016. Conventional drinking water treatment and direct biofiltration for the removal of pharmaceuticals and artificial sweeteners: A pilot-scale approach. *Science of The Total Environment* 544, 10-17.<https://doi.org/10.1016/j.scitotenv.2015.11.145>

Mendes, C., Laignier, E., Brigagão, M., Luccas, P., Tarley, C., 2010. A simple turbidimetric flow injection system for saccharin determination in sweetener products. *Chemical Papers* 64(3), 285-293.<https://doi.org/10.2478/s11696-010-0009-7>

Miklos, D.B., Remy, C., Jekel, M., Linden, K.G., Drewes, J.E., Hübner, U., 2018. Evaluation of advanced oxidation processes for water and wastewater treatment – A critical review. *Water Research* 139, 118-131.<https://doi.org/10.1016/j.watres.2018.03.042>

Ochsenbein, U., Zeh, M., Berset, J.-D., 2008. Comparing solid phase extraction and direct injection for the analysis of ultra-trace levels of relevant explosives in lake water and tributaries using liquid chromatography–electrospray tandem mass spectrometry. *Chemosphere* 72(6), 974-980.<https://doi.org/10.1016/j.chemosphere.2008.03.004>

Oliveira, V., Oliveira, V., Oliveira, T., Damasceno, A., Silva, C., Medeiros, S., Soares, B., Carneiro, F., Aguiar, R., Islam, M., Amélia, A., Peron, A., Sousa, J., 2017. Evaluation of cytotoxic and mutagenic

effects of two artificial sweeteners using eukaryotic test systems. *African Journal of Biotechnology* 16.10.5897/AJB2016.15695

Ordóñez, E.Y., Quintana, J.B., Rodil, R., Cela, R., 2013. Determination of artificial sweeteners in sewage sludge samples using pressurised liquid extraction and liquid chromatography–tandem mass spectrometry. *Journal of Chromatography A* 1320, 10-16.<https://doi.org/10.1016/j.chroma.2013.10.049>

Ordóñez, E.Y., Quintana, J.B., Rodil, R., Cela, R., 2012. Determination of artificial sweeteners in water samples by solid-phase extraction and liquid chromatography–tandem mass spectrometry. *Journal of Chromatography A* 1256, 197-205.<https://doi.org/10.1016/j.chroma.2012.07.073>

Oser, B.L., 1985. Highlights in the history of saccharin toxicology. *Food and Chemical Toxicology* 23(4), 535-542.[https://doi.org/10.1016/0278-6915\(85\)90148-6](https://doi.org/10.1016/0278-6915(85)90148-6)

Pak, G., Salcedo, D.E., Lee, H., Oh, J., Maeng, S.K., Song, K.G., Hong, S.W., Kim, H.-C., Chandran, K., Kim, S., 2016. Comparison of antibiotic resistance removal efficiencies using ozone disinfection under different pH and suspended solids and humic substance concentrations. *Environmental Science & Technology* 50(14), 7590-7600.10.1021/acs.est.6b01340

Pal, A., He, Y., Jekel, M., Reinhard, M., Gin, K.Y.-H., 2014. Emerging contaminants of public health significance as water quality indicator compounds in the urban water cycle. *Environment International* 71, 46-62.<https://doi.org/10.1016/j.envint.2014.05.025>

Paruch, L., Paruch, A.M., Blankenberg, A.-G.B., Haarstad, K., Mæhlum, T., 2017. Norwegian study on microbial source tracking for water quality control and pollution removal in constructed wetland treating catchment run-off. *Water Science and Technology* 76(5), 1158-1166.10.2166/wst.2017.303

Perkola, N., Sainio, P., 2014. Quantification of four artificial sweeteners in Finnish surface waters with isotope-dilution mass spectrometry. *Environmental Pollution* 184, 391-396.<https://doi.org/10.1016/j.envpol.2013.09.017>

Perkola, N., Vaalgamaa, S., Jernberg, J., Vähätalo, A.V., 2016. Degradation of artificial sweeteners via direct and indirect photochemical reactions. *Environmental Science and Pollution Research* 23(13), 13288-13297.10.1007/s11356-016-6489-4

Phan, H.V., Wickham, R., Xie, S., McDonald, J.A., Khan, S.J., Ngo, H.H., Guo, W., Nghiem, L.D., 2018. The fate of trace organic contaminants during anaerobic digestion of primary sludge: A pilot scale study. *Bioresource Technology* 256, 384-390.<https://doi.org/10.1016/j.biortech.2018.02.040>

Qin, X., 2012. Food additives: possible cause for recent remarkable increase of inflammatory bowel disease in children. *Journal of Pediatric Gastroenterology and Nutrition* 54(4), 564.10.1097/MPG.0b013e3182464b0e

Qin, X., 2016. The possible link between artificial sweeteners such as saccharin and sucralose and inflammatory bowel disease deserves further study. *Inflammatory bowel diseases* 22(6), E17.10.1097/mib.0000000000000760

Qin, X., 2019. Impaired inactivation of digestive proteases in lower gut due to inhibition of gut bacteria by food additives such as saccharin and sucralose as main cause of inflammatory bowel disease: a two-decades-long hypothesis warrants testing. *Inflammatory Bowel Diseases* 25(11), e141-e141.10.1093/ibd/izz187

Qu, J., Chen, X., Zhou, J., Li, H., Mai, W., 2019. Treatment of real sodium saccharin wastewater using multistage contact oxidation reactor and microbial community analysis. *Bioresource Technology* 289, 121714.<https://doi.org/10.1016/j.biortech.2019.121714>

Rashidi, A.M., Amadeh, A., 2009. The effect of saccharin addition and bath temperature on the grain size of nanocrystalline nickel coatings. *Surface and Coatings Technology* 204(3), 353-

358.<https://doi.org/10.1016/j.surfcoat.2009.07.036>

Richards, S., Withers, P.J.A., Paterson, E., McRoberts, C.W., Stutter, M., 2017. Removal and attenuation of sewage effluent combined tracer signals of phosphorus, caffeine and saccharin in soil. *Environmental Pollution* 223, 277-285.<https://doi.org/10.1016/j.envpol.2017.01.024>

Richter, D., Dünnbier, U., Massmann, G., Pekdeger, A., 2007. Quantitative determination of three sulfonamides in environmental water samples using liquid chromatography coupled to electrospray tandem mass spectrometry. *Journal of Chromatography A* 1157(1), 115-121.<https://doi.org/10.1016/j.chroma.2007.04.042>

Rostvall, A., Zhang, W., Dürig, W., Renman, G., Wiberg, K., Ahrens, L., Gago-Ferrero, P., 2018. Removal of pharmaceuticals, perfluoroalkyl substances and other micropollutants from wastewater using lignite, Xylit, sand, granular activated carbon (GAC) and GAC+Polonite® in column tests – Role of physicochemical properties. *Water Research* 137, 97-106.<https://doi.org/10.1016/j.watres.2018.03.008>

Roy, J.W., Van Stempvoort, D.R., Bickerton, G., 2014. Artificial sweeteners as potential tracers of municipal landfill leachate. *Environmental Pollution* 184, 89-93.<https://doi.org/10.1016/j.envpol.2013.08.021>

Russo, D., Cochran, K.H., Westerman, D., Li Puma, G., Marotta, R., Andreozzi, R., Richardson, S.D., 2020. Ultrafast photodegradation of isoxazole and isothiazolinones by UV254 and UV254/H<sub>2</sub>O<sub>2</sub> photolysis in a microcapillary reactor. *Water Research* 169, 115203.<https://doi.org/10.1016/j.watres.2019.115203>

Sang, Z., Jiang, Y., Tsoi, Y.-K., Leung, K.S.-Y., 2014. Evaluating the environmental impact of artificial sweeteners: A study of their distributions, photodegradation and toxicities. *Water Research* 52, 260-274.<https://doi.org/10.1016/j.watres.2013.11.002>

Sastry, S.K., Datta, A.K., Worobo, R.W., 2000. Ultraviolet Light. *Journal of Food Science* 65(s8), 90-92.10.1111/j.1750-3841.2000.tb00623.x

Scheurer, M., Brauch, H.-J., Lange, F.T., 2009. Analysis and occurrence of seven artificial sweeteners in German waste water and surface water and in soil aquifer treatment (SAT). *Analytical and Bioanalytical Chemistry* 394(6), 1585-1594.10.1007/s00216-009-2881-y

Scheurer, M., Brauch, H.-J., Lange, F.T., 2014. Transformation Products of Artificial Sweeteners. *Transformation products of emerging contaminants in the environment*, 525-544.doi:10.1002/9781118339558.ch1710.1002/9781118339558.ch17

Scheurer, M., Storck, F.R., Brauch, H.-J., Lange, F.T., 2010. Performance of conventional multi-barrier drinking water treatment plants for the removal of four artificial sweeteners. *Water Research* 44(12), 3573-3584.<https://doi.org/10.1016/j.watres.2010.04.005>

Schleheck, D., Cook, A.M., 2003. Saccharin as a sole source of carbon and energy for *Sphingomonas xenophaga* SKN. *Archives of Microbiology* 179(3), 191-196.10.1007/s00203-002-0515-2

Seitz, W., Winzenbacher, R., 2017. A survey on trace organic chemicals in a German water protection area and the proposal of relevant indicators for anthropogenic influences. *Environmental Monitoring and Assessment* 189(6), 244.10.1007/s10661-017-5953-z

Seo, P.W., Khan, N.A., Hasan, Z., Jhung, S.H., 2016. Adsorptive removal of artificial sweeteners from water using metal–organic frameworks functionalized with urea or melamine. *ACS Applied Materials & Interfaces* 8(43), 29799-29807.10.1021/acsami.6b11115

Shao, Y., Pang, Z., Wang, L., Liu, X., 2019. Efficient degradation of acesulfame by ozone/peroxymonosulfate advanced oxidation process. *Molecules* 24(16), 2874.<https://doi.org/10.3390/molecules24162874>

- Shreve, M.J., Brennan, R.A., 2019. Trace organic contaminant removal in six full-scale integrated fixed-film activated sludge (IFAS) systems treating municipal wastewater. *Water Research* 151, 318-331. <https://doi.org/10.1016/j.watres.2018.12.042>
- Slavič, A., Kalčikova, G., Jemec Kokalj, A., Žgajnar Gotvajn, A., 2018. Biodegradation of the artificial sweetener saccharin in surface waters and groundwaters. *Acta hydrotechnica* 31(55), 157-165. <https://doi.org/10.15292/acta.hydro.2018.10>
- Snider, D.M., Roy, J.W., Robertson, W.D., Garda, D.I., Spoelstra, J., 2017. Concentrations of artificial sweeteners and their ratios with nutrients in septic system wastewater. *Groundwater Monitoring & Remediation* 37(3), 94-102. [10.1111/gwmm.12229](https://doi.org/10.1111/gwmm.12229)
- Soh, L., Connors, K.A., Brooks, B.W., Zimmerman, J., 2011. Fate of sucralose through environmental and water treatment processes and impact on plant indicator species. *Environmental Science & Technology* 45(4), 1363-1369. [10.1021/es102719d](https://doi.org/10.1021/es102719d)
- Song, J.Y., Bhadra, B.N., Khan, N.A., Jung, S.H., 2018. Adsorptive removal of artificial sweeteners from water using porous carbons derived from metal azolate framework-6. *Microporous and Mesoporous Materials* 260, 1-8. <https://doi.org/10.1016/j.micromeso.2017.10.021>
- Spoelstra, J., Roy, J.W., Brown, S.J., 2016. Syringe-tip filters may contain the artificial sweetener saccharin. *Groundwater Monitoring & Remediation* 36(4), 71-74. [10.1111/gwmm.12183](https://doi.org/10.1111/gwmm.12183)
- Spoelstra, J., Schiff, S.L., Brown, S.J., 2013. Artificial sweeteners in a large Canadian river reflect human consumption in the watershed. *PloS one* 8(12). [10.1371/journal.pone.0082706](https://doi.org/10.1371/journal.pone.0082706)
- Stefan, M.I., 2017. *Advanced oxidation processes for water treatment: fundamentals and applications*. IWA publishing.
- Stefania, G.A., Rotiroti, M., Buerge, I.J., Zanotti, C., Nava, V., Leoni, B., Fumagalli, L., Bonomi, T., 2019. Identification of groundwater pollution sources in a landfill site using artificial sweeteners, multivariate analysis and transport modeling. *Waste Management* 95, 116-128. <https://doi.org/10.1016/j.wasman.2019.06.010>
- Subedi, B., Balakrishna, K., Sinha, R.K., Yamashita, N., Balasubramanian, V.G., Kannan, K., 2015. Mass loading and removal of pharmaceuticals and personal care products, including psychoactive and illicit drugs and artificial sweeteners, in five sewage treatment plants in India. *Journal of Environmental Chemical Engineering* 3(4, Part A), 2882-2891. <https://doi.org/10.1016/j.jece.2015.09.031>
- Subedi, B., Kannan, K., 2014. Fate of artificial sweeteners in wastewater treatment plants in New York State, U.S.A. *Environmental Science & Technology* 48(23), 13668-13674. [10.1021/es504769c](https://doi.org/10.1021/es504769c)
- Subedi, B., Lee, S., Moon, H.-B., Kannan, K., 2014. Emission of artificial sweeteners, select pharmaceuticals, and personal care products through sewage sludge from wastewater treatment plants in Korea. *Environment International* 68, 33-40. <https://doi.org/10.1016/j.envint.2014.03.006>
- Torres, C.I., Ramakrishna, S., Chiu, C.-A., Nelson, K.G., Westerhoff, P., Krajmalnik-Brown, R., 2011. Fate of sucralose during wastewater treatment. *Environmental Engineering Science* 28(5), 325-331. [10.1089/ees.2010.0227](https://doi.org/10.1089/ees.2010.0227)
- Toth, J.E., Rickman, K.A., Venter, A.R., Kiddle, J.J., Mezyk, S.P., 2012. Reaction kinetics and efficiencies for the hydroxyl and sulfate radical based oxidation of artificial sweeteners in water. *The Journal of Physical Chemistry A* 116(40), 9819-9824. [10.1021/jp3047246](https://doi.org/10.1021/jp3047246)
- Tran, N.H., Gan, J., Nguyen, V.T., Chen, H., You, L., Duarah, A., Zhang, L., Gin, K.Y.-H., 2015. Sorption and biodegradation of artificial sweeteners in activated sludge processes. *Bioresource Technology* 197, 329-338. <https://doi.org/10.1016/j.biortech.2015.08.083>
- Tran, N.H., Hu, J., Li, J., Ong, S.L., 2014a. Suitability of artificial sweeteners as indicators of raw

wastewater contamination in surface water and groundwater. *Water Research* 48, 443-456.<https://doi.org/10.1016/j.watres.2013.09.053>

Tran, N.H., Nguyen, V.T., Urase, T., Ngo, H.H., 2014b. Role of nitrification in the biodegradation of selected artificial sweetening agents in biological wastewater treatment process. *Bioresource Technology* 161, 40-46.<https://doi.org/10.1016/j.biortech.2014.02.116>

Tran, N.H., Reinhard, M., Gin, K.Y.-H., 2018. Occurrence and fate of emerging contaminants in municipal wastewater treatment plants from different geographical regions-a review. *Water Research* 133, 182-207.<https://doi.org/10.1016/j.watres.2017.12.029>

Tran, N.H., Reinhard, M., Khan, E., Chen, H., Nguyen, V.T., Li, Y., Goh, S.G., Nguyen, Q.B., Saedi, N., Gin, K.Y.-H., 2019. Emerging contaminants in wastewater, stormwater runoff, and surface water: Application as chemical markers for diffuse sources. *Science of The Total Environment* 676, 252-267.<https://doi.org/10.1016/j.scitotenv.2019.04.160>

Uçar, A., Yılmaz, S., 2015. Saccharin genotoxicity and carcinogenicity: a review. *Advances in Food Sciences* 37, 138-142

Van Stempvoort, D.R., Robertson, W.D., Brown, S.J., 2011a. Artificial sweeteners in a large septic plume. *Groundwater Monitoring & Remediation* 31(4), 95-102.[10.1111/j.1745-6592.2011.01353.x](https://doi.org/10.1111/j.1745-6592.2011.01353.x)

Van Stempvoort, D.R., Roy, J.W., Brown, S.J., Bickerton, G., 2011b. Artificial sweeteners as potential tracers in groundwater in urban environments. *Journal of Hydrology* 401(1), 126-133.<https://doi.org/10.1016/j.jhydrol.2011.02.013>

Van Stempvoort, D.R., Roy, J.W., Grabuski, J., Brown, S.J., Bickerton, G., Sverko, E., 2013. An artificial sweetener and pharmaceutical compounds as co-tracers of urban wastewater in groundwater. *Science of The Total Environment* 461-462, 348-359.<https://doi.org/10.1016/j.scitotenv.2013.05.001>

Vymazal, J., Dvořáková Březinová, T., 2016. Removal of saccharin from municipal sewage: The first results from constructed wetlands. *Chemical Engineering Journal* 306, 1067-1070.<https://doi.org/10.1016/j.cej.2016.08.043>

Wang, J., Wang, S., 2019. Preparation, modification and environmental application of biochar: A review. *Journal of Cleaner Production* 227, 1002-1022.<https://doi.org/10.1016/j.jclepro.2019.04.282>

Wang, L., Yang, L., Li, Y., Zhang, Y., Ma, X., Ye, Z., 2010. Study on adsorption mechanism of Pb(II) and Cu(II) in aqueous solution using PS-EDTA resin. *Chemical Engineering Journal* 163(3), 364-372.<https://doi.org/10.1016/j.cej.2010.08.017>

Wang, S., 2008. A Comparative study of Fenton and Fenton-like reaction kinetics in decolourisation of wastewater. *Dyes and Pigments* 76(3), 714-720.<https://doi.org/10.1016/j.dyepig.2007.01.012>

Wang, Z., Nguyen Song Thuy Thuy, G., Srivastava, V., Ambat, I., Sillanpää, M., 2019. Photocatalytic degradation of an artificial sweetener (Acesulfame-K) from synthetic wastewater under UV-LED controlled illumination. *Process Safety and Environmental Protection* 123, 206-214.<https://doi.org/10.1016/j.psep.2019.01.018>

Watanabe, E., Nascimento, A.P., Guerreiro-Tanomaru, J.M., Razaboni, A.M., de Andrade, D., Tanomaru-Filho, M., 2015. Antiseptic mouthwashes: in vitro antibacterial activity. *Acta Odontológica Latinoamericana* 28(2), 180-184

Watanabe, Y., Bach, L.T., Van Dinh, P., Prudente, M., Aguja, S., Phay, N., Nakata, H., 2016. ubiquitous detection of artificial sweeteners and iodinated x-ray contrast media in aquatic environmental and wastewater treatment plant samples from vietnam, the philippines, and myanmar. *Archives of Environmental Contamination and Toxicology* 70(4), 671-681.[10.1007/s00244-015-0220-1](https://doi.org/10.1007/s00244-015-0220-1)

Weihrauch, M., Diehl, V., 2004. Artificial sweeteners—do they bear a carcinogenic risk? *Annals of*



Oncology 15(10), 1460-1465

Xiao, J., Xie, Y., Cao, H., 2015. Organic pollutants removal in wastewater by heterogeneous photocatalytic ozonation. *Chemosphere* 121, 1-17.<https://doi.org/10.1016/j.chemosphere.2014.10.072>

Xu, Y., Lin, Z., Zhang, H., 2016. Mineralization of sucralose by UV-based advanced oxidation processes: UV/PDS versus UV/H<sub>2</sub>O<sub>2</sub>. *Chemical Engineering Journal* 285, 392-401.<https://doi.org/10.1016/j.cej.2015.09.091>

Yang, Y.-Y., Liu, W.-R., Liu, Y.-S., Zhao, J.-L., Zhang, Q.-Q., Zhang, M., Zhang, J.-N., Jiang, Y.-X., Zhang, L.-J., Ying, G.-G., 2017. Suitability of pharmaceuticals and personal care products (PPCPs) and artificial sweeteners (ASs) as wastewater indicators in the Pearl River Delta, South China. *Science of The Total Environment* 590-591, 611-619.<https://doi.org/10.1016/j.scitotenv.2017.03.001>

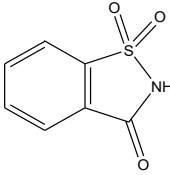
Zelinski, D.W., dos Santos, T.P.M., Takashina, T.A., Leifeld, V., Igarashi-Mafra, L., 2018. Photocatalytic degradation of emerging contaminants: artificial sweeteners. *Water, Air, & Soil Pollution* 229(6), 207.10.1007/s11270-018-3856-4

Zhao, Y.-G., Cai, M.-Q., Chen, X.-H., Pan, S.-D., Yao, S.-S., Jin, M.-C., 2013. Analysis of nine food additives in wine by dispersive solid-phase extraction and reversed-phase high performance liquid chromatography. *Food Research International* 52(1), 350-358.<https://doi.org/10.1016/j.foodres.2013.03.038>

Zhu, Y., Guo, Y., Ye, M., James, F.S., 2005. Separation and simultaneous determination of four artificial sweeteners in food and beverages by ion chromatography. *Journal of Chromatography A* 1085(1), 143-146.<https://doi.org/10.1016/j.chroma.2004.12.042>

Zygler, A., Wasik, A., Namieśnik, J., 2009. Analytical methodologies for determination of artificial sweeteners in foodstuffs. *TrAC Trends in Analytical Chemistry* 28(9), 1082-1102.<https://doi.org/10.1016/j.trac.2009.06.008>

**Table****Table 1** Physicochemical properties of saccharin

Name	Structure	Properties	Acceptable daily intake
Saccharin (1,1-dioxo-1,2-benzothiazol-3-one; Benzoic sulfimide)		CAS No.: 81-07-2; Chemical formula: C <sub>7</sub> H <sub>5</sub> NO <sub>3</sub> S; Molecular weight: 183.18 g/mol; pK <sub>a</sub> : 2.2; log K <sub>ow</sub> : 0.91; Water solubility: 4 g/L	5 mg/kg body weight (sodium salt); 3.8 mg/kg body weight (free acid)

**Table 2.** Analytical techniques with limits of detection (LODs) and limits of quantitation (LOQs) for SAC detection in different aqueous samples.

Analytical technique	Water matrix <sup>a</sup>	LODs (µg/L)	LOQs (µg/L)	Reference
High-pressure LC	Animal feed	N/A <sup>b</sup> (low to 10 ppm)	N/A	(Holder and Bowman, 1980)
	Human urine	N/A (low to 10 ppm)	N/A	
	Wastewater	N/A (low to 0.1 ppm)	N/A	
LC-tandem mass spectrometry (MS/MS) (LC-MS/MS)	Surface water	N/A	0.25	(Ordóñez et al., 2012)
	Wastewater	N/A	0.50	
	Tap water/Wastewater	0.24 (instrument)	0.80(instrument)	(Kokotou and Thomaidis, 2013)
	River water	0.001-0.04	N/A	
	Wastewater	0.01-0.5	N/A	(Arbeláez et al., 2015b)
	Groundwater	0.3	N/A	
	LC with electrospray ionization source equipped MS (LC-ESI-MS)	River/Wastewater	1.5×10 <sup>-5</sup> -0.023 (instrument)	
LC time-of-flight MS (LC-TOF-MS)	Soft drink and liquid (no extraction)	100 (instrument)	1000 (instrument)	(Watanabe et al., 2016)
	Ground/surface/waste water	0.5 (instrument)	5 (instrument)	
LC-quadrupole(Q)-TOF-MS	Surface/waste water	N/A	N/A	(Hernández et al., 2015)
	Artificial wastewater(filtration)	N/A	N/A	(Davididou et al., 2019)
HPLC-MS/MS	Ground/river/tap/waste water	N/A	0.01	(Berset and Ochsenbein, 2012)
UFLC-Q-MS/MS	Red wine	1.5	5	(Chen et al., 2012)
Ion-pair HPLC triple quadrupole-MS (IP- HPLC-TQ-MS)	River water	0.0003	0.0008	(Gan et al., 2013b)
	Tap water	0.0003	0.0011	
	Sea water	0.0005	0.0018	
LC-isotope-dilution MS (LC-IDMS)	Surface water	0.004	0.025	(Perkola and Sainio, 2014)
LC-high resolution MS(LC-HR-MS)	Artificial wastewater(filtration)	0.005	0.1	(Davididou et al., 2018)
RP-HPLC-Q-TOF-MS	Wastewater	N/A	N/A	(Kempińska and Kot-Wasik,

Analytical technique	Water matrix <sup>a</sup>	LODs (µg/L)	LOQs (µg/L)	Reference
				2018)
FIA coupled a monolithic column	Artificial wastewater	N/A	N/A	(Fatibello-Filho et al., 1994)
	Artificial wastewater	1.5×10 <sup>-5</sup> mol/L	N/A	(Fatibello-Filho and Aniceto, 1999)
	Liquid sweetener products	N/A	2.4×10 <sup>6</sup>	(Mendes et al., 2010)
CE with capacitively coupled contactless conductivity detection (C <sup>4</sup> D)	Soft drink (filtration)	N/A	10	(Frazier et al., 2000)
	Soft drink (dilution)	1500	4900	(Bergamo et al., 2011)
IC with suppressed conductivity detector	Carbonated cola drink(ultrasonic+dilution)	45	N/A	(Zhu et al., 2005)
	Fruit juice drink (dilution)			
	Preserved fruit (ultrasonic+dilution)			
TLC	Toddy	0.1µg/spot (9 µg/cm <sup>2</sup> )	N/A	(Mali et al., 2005)

**a.** samples in this column all are extracted except those indicated otherwise; **b.** N/A: not applicable.

**Table 3** Research information on saccharin occurrence in the aqueous environment including wastewater and sludge in WWTPs and wetlands, surface water, groundwater, runoff, tap water (drinking water), and sea water.

Water Matrix	Location	Year	Range/Concentration (ng/L)	Determination Method	Reference
Wastewater	Karlsruhe, German (WWTPs)	N/A <sup>a</sup>	34,000-50,000 (in <sup>b</sup> ); 400 (out <sup>c</sup> )	HPLC-ESI-TQ-MS	(Scheurer et al., 2009)
	Zurich, Switzerland (WWTPs)	Jun, Oct 2008	3900-18,000 (in); <LOD-3200 (out)	LC-MS/MS	(Buerge et al., 2009)
	US (WWTPs)	N/A	<LOD-5000 (out)	LC-TOF-MS	(Ferrer and Thurman, 2010)
	Zurich, Switzerland (WWTPs)	N/A	4000-18,000 (in); <100-3000 (out)	LC-MS/MS	(Buerge et al., 2011)
	Switzerland (WWTPs)	May 9-10, 2011	86-16,201 (out)	HPLC-MS/MS	(Berset and Ochsenbein, 2012)
	NW Spain (WWTPs)	Sep 2011	18,400-22,300 (in); 7100-9100 (out)	LC-ESI-TQ-MS	(Ordóñez et al., 2012)
	Athens, Greece (WWTPs)	N/A	15,000-46,000 (in); <LOD-270 (out)	LC-ESI-MS/MS	(Kokotou and Thomaidis, 2013)
	Tianjin, China (WWTPs)	N/A	~10,300 (in); ~242 (out)	IPLC-ESI-TQ-MS/MS	(Gan et al., 2013b)
	Tianjin, China (WWTPs+Wetlands)	Jun 28, 2011; Nov 6, 2011	WWTPs: 7200-9100 (in); 270-280 (out) Wetlands: 140-560	LC-TQ-MS/MS	(Gan et al., 2013a)
	Singapore (WWTPs)	N/A	10 <sup>1</sup> -10 <sup>6</sup> (in)	HPLC-ID-MS/MS	(Tran et al., 2014a)

Water Matrix	Location	Year	Range/Concentration (ng/L)	Determination Method	Reference
Wastewater	Tianjin, China (WWTPs)	Dec 13-26, 2012; Sep 11-24, 2013	210 ±160 (AVG ±SD) <sup>d</sup>	LC-MS/MS	(Gan et al., 2014)
	Albany, USA (WWTPs)	Jul 12-18, 2013	1860-25,100 (in); 130-15,200 (out)	HPLC-MS/MS	(Subedi and Kannan, 2014)
	Tarragona, Spain (WWTPs)	N/A	<LOQ-155,000 (in); <LOQ-500 (out)	HPLC-ESI-MS/MS	(Arbeláez et al., 2015b)
	India (WWTPs)	Jul-Aug 2012	303,000 (in, highest M <sup>e</sup> ); 12,120 (out, removal rate 96%)	HPLC-ESI-MS/MS	(Subedi et al., 2015)
	Singapore (Water reclamation plant)	Feb-Jul 2013	9310-18,797 (in); <LOD (out)	HPLC-MS/MS	(Tran et al., 2015)
	Hanoi, Vietnam (WWTPs)	Oct, Dec 2013	7600-13,000 (in); 23-36 (out)	LC-ESI-MS	(Watanabe et al., 2016)
	Czech Republic (Constructed wetland)	Oct 2013-Nov 2015	6,200-87,000 (in); 2600-37,000 (out, AVG removal rate 42.4% )	UHPLC-ESI-TQ-MS/MS	(Vymazal and Dvořáková Březinová, 2016)
	South German (WWTPs)	2012-2014	14,000 (in, highest M)	HPLC-ESI-MS/MS	(Seitz and Winzenbacher, 2017)
	Ontario (Septic tanks)	N/A	6100-72,000 (single dwelling); 2200-63,000 (multiple dwelling)	IC-ESI-MS/MS	(Snider et al., 2017)
	Pearl River Delta region, South China (WWTPs)	May-Oct 2015	8230 (in, M); <823 (out, removal rate > 90%)	UHPLC-ESI-MS-MS	(Yang et al., 2017)
Tianjin, China (Farm WWTPs)	Aug 2015-May 2016	781,000 ±9000 (in); 11,100 ±100 (primary out); 5640 ±370 (out)	LC-TQ-MS/MS	(Baalbaki et al., 2017)	

Water Matrix	Location	Year	Range/Concentration (ng/L)	Determination Method	Reference
Wastewater	Nanjing/Wuxi, China (WWTPs)	Oct 15, 2015; Jan 15, Apr 15, Jul 15, 2016	430-27,340 (in); 20-240 (out)	UPLC-ESI-TQ-S MS/MS	(Li et al., 2018b)
	Tarragona & Reus, Spain (WWTPs)	N/A	4700-17,900 (out)	HPLC-MS/MS	(Lakade et al., 2018)
	N/A (WWTPs)	N/A	4408-52,962 (in); <LOQ-5663 (out)	HPLC-MS/MS	(Tran et al., 2019)
Sludge	Zurich, Switzerland (Farm WWTPs)	N/A	liquid manure: $3 \times 10^5$ - $1.2 \times 10^7$ ; manure fermentation: $300$ - $5 \times 10^5$ ; sludge: $1 \times 10^4$ - $1.6 \times 10^4$	LC-MS/MS	(Buerge et al., 2011)
	NW Spain (WWTPs)	N/A	141-255 ng/g dwS <sup>f</sup> (primary); <LOD-124 ng/g dwS (secondary)	LC-ESI-MS/MS	(Ordoñez et al., 2013)
	South Korea (WWTPs, Mixed: Industrial + Domestic)	Jul-Oct 2011	11.4-55.4 ng/g dwS (Industrial); 7.08-3240 ng/g dwS (Domestic); 11.6-54.0 ng/g dwS (Mixed);	HPLC-ESI-TQ-MS/MS	(Subedi et al., 2014)
	Albany, USA (WWTPs)	Jul 12-18, 2013	<LOD-388,000	HPLC-MS/MS	(Subedi and Kannan, 2014)
	Catalonia, Spain (WWTPs)	consecutive 3 months	105-591 ng/g dwS	HPLC-ESI-TQ-MS/MS	(Arbeláez et al., 2015a)
	India (WWTPs)	Jul-Aug 2012	18,700 ng/g dwS	HPLC-ESI-MS/MS	(Subedi et al., 2015)
Surface water	Swiss Midland region	Feb-Mar 2008	<LOD-2800	LC-MS/MS	(Buerge et al., 2009)
	Switzerland	May3,16-19, 2011	<LOQ-310	HPLC-MS/MS	(Berset and Ochsenbein, 2012)
	NW Spain	Sep 2011	<LOD-19,700	LC-ESI-TQ-MS	(Ordóñez et al., 2012)
	Tianjin, China	N/A	193-746	IPLC-ESI-TQ-MS/MS	(Gan et al., 2013b)
	Tianjin, China	Jul 9-15, 2011,	210-1100	LC-TQ-MS/MS	(Gan et al., 2013a)

<b>Water Matrix</b>	<b>Location</b>	<b>Year</b>	<b>Range/Concentration (ng/L)</b>	<b>Determination Method</b>	<b>Reference</b>
<b>Surface water</b>	Ontario, Canada	Jun 14, Sep 5, 2007; Apr 24, 2009	<LOD-7200	IC-ESI-MS/MS	(Spoelstra et al., 2013)
	Singapore	N/A	<LOD-3210	HPLC-ID-MS/MS	(Tran et al., 2014a)
	Finland	Jun, Oct 2011; May, Jun 2012	<25-490	UPLC-ESI-ID-MS/MS	(Perkola and Sainio, 2014)
	Hong Kong, China	Jul 22, 2011; Jan 17, 2012	250	UHPLC-ESI-TQ-MS/MS	(Sang et al., 2014)
	Tianjin, China	Dec 13-26, 2012; Sep 11-24, 2013	120-62,700	LC-MS/MS	(Gan et al., 2014)
	Chennai, India	Jul - Aug 2012	419,000	HPLC-ESI-MS/MS	(Subedi et al., 2015)
	Hanoi/Haiphong Vietnam	Oct, Dec 2013	0.092-17,000	LC-ESI-MS	(Watanabe et al., 2016)
	Manila, The Philippines	Jun 2014	290-9600	LC-ESI-MS	(Watanabe et al., 2016)
	Pathein/Yangon, Myanmar	Dec 2014	<LOD-71	LC-ESI-MS	(Watanabe et al., 2016)
	Barbados, West Indies	Feb, Jun 2013	<LOQ-488±46	LC-ESI-MS/MS	(Edwards et al., 2017)
	River Ebro	N/A	100-200	HPLC-MS/MS	(Lakade et al., 2018)
	Northern Italy	Mar 2017	<LOQ-530	HPLC-MS/MS	(Stefania et al., 2019)
	N/A	Apr 2015-Dec 2016	<LOQ-5663	HPLC-MS/MS	(Tran et al., 2019)
<b>Groundwater</b>	Mediterranean country, German	N/A	<LOD-<400	HPLC-ESI-TQ-MS	(Scheurer et al., 2009)
	Zurich, Swizerland	Aug, Nov 2008	<LOD	LC-MS/MS	(Buerge et al., 2009)
	Canada (Lagoon)	N/A	<LOD-10,300	IC-ESI-TQ-MS/MS (isotope-labled)	(Van Stempvoort et al., 2011b)
	Ontario, Canada	Sep 2008; Jun, Oct 2009; Mar-Apr 2010	21-40,000	IC-ESI-TQ-MS/MS	(Van Stempvoort et al., 2011a)



<b>Water Matrix</b>	<b>Location</b>	<b>Year</b>	<b>Range/Concentration (ng/L)</b>	<b>Determination Method</b>	<b>Reference</b>
	Switzerland	May 23 -24 2011	<LOQ-15	HPLC-MS/MS	(Berset and Ochsenbein, 2012)
	Tianjin, China	N/A	11.3-80.0	IPLC-ESI-TQ-MS/MS	(Gan et al., 2013b)
<b>Groundwater</b>	Barrie/Jasper, Canada	Sep-Nov 2010	<LOD-35	LC-MS/MS	(Van Stempvoort et al., 2013)
	Tianjin, China	Jul 2011	52-63	LC-TQ-MS/MS	(Gan et al., 2013a)
	Singapore	N/A	<LOD-780	HPLC-ID-MS/MS	(Tran et al., 2014a)
	Canada (Landfill monitoring well)	N/A	<LOQ-250,000	IC-ESI-TQ-MS/MS	(Roy et al., 2014)
	Halong, Vietnam	Oct, Dec 2013	<LOD-13	LC-ESI-MS	(Watanabe et al., 2016)
	Barbados, West Indies	Feb, Jun 2013	2.7±0.2 - 9.6±2.1	LC-ESI-MS/MS	(Edwards et al., 2019)
	Northern Italy	Mar 2017	<LOQ -680	HPLC-MS/MS	(Stefania et al., 2019)
<b>Runoff</b>	Gryteland stream, Norway	Nov 2014; Feb, Jun, Sep, Dec 2015; Mar 2016	<LOQ-80 (in); <LOQ-74 (out)	UHPLC-TQ-MS/MS	(Paruch et al., 2017)
	N/A	Apr 2015-Dec 2016	45-9125 (urban) 17-1028 (Agricultural)	HPLC-MS/MS	(Tran et al., 2019)
<b>Tap water</b>	Tianjin, China	N/A	47.5-65.1	IPLC-ESI-TQ-MS/MS	(Gan et al., 2013b)
	Tianjin, China	Jul 2011	Up to 110	LC-TQ-MS/MS	(Gan et al., 2013a)
	Canada	N/A	<LOD-350	IC-ESI-MS/MS	(Spoelstra et al., 2013)
<b>Sea water</b>	Tianjin, China	N/A	99.7-249	IPLC-ESI-TQ-MS/MS	(Gan et al., 2013b)
	Hong Kong, China	Jul 22, 2011; Jan 17, 2012	<LOD-300	UHPLC-ESI-TQ-MS/MS	(Sang et al., 2014)
	Western basin, the Mediterranean Sea	Autumn 2014	0.49-5.23	UPLC-MS/MS; HPLC-MS/MS	(Brumovský et al., 2017)

**a.** N/A: not applicable; **b.** in: inflow/influent/inlet/raw/untreated water in related references; **c.** out: outflow/effluent/outlet/treated water in related references; **d.** AVG±SD: average concentration ± standard deviation; **e.** M: median concentration; **f.** ng/g dwS: ng/g dry weight sludge.

**Table 4.**Physical, chemical, and biological processes/techniques for SAC treatment

Treatment technique		Main mechanism	SAC optimal removal performance	Reference
Physical	Activated carbon (AC)	Adsorption	<b>1. AC</b> (lab-scale) 40% (only AC adsorption 16 h); 75% (ultrasonication 180 min + AC adsorption, 16 h)	(Bernardo et al., 2006)
		Adsorption + Biodegradation (main)	<b>2. Granular AC (GAC)</b> (lab-scale) 0.4 mg SAC/g GAC	(Li et al., 2018b)
		Adsorption	<b>3. Powdered AC (PAC)</b> (pilot-scale) 33-54% (fluidized bed)	(Mailler et al., 2015)
	<b>Biochar(BC)</b>	Adsorption ( $\pi$ - $\pi$ stacking)	Partial removal (lab-scale)	(Liu et al., 2019)
	<b>Filtration</b>	Adsorption + Biodegradation (main)	<b>1. Bank filtration</b> (sampling study) Nearly 100% (by soil) <b>2. Sand filtration</b> (sampling study) 14.5% (by mainly quartz sand and anthracites)	(Buerge et al., 2011) (Gan et al., 2012)
	<b>Metal-organic framework (MOF)</b>	Adsorption (H-bonding)	<b>1. Maximum adsorption capacity of MIL</b> (name for a specially prepared MOF) (lab-scale) 53.4 mg SAC / g MIL-101 70.1 mg SAC / g melamine-MIL-101 86.4 mg SAC / g urea-MIL-101 18.7 mg SAC / g O <sub>2</sub> N-MIL-101 <b>2. Maximum adsorption capacity of metal azolate framework-6 derived porous carbons (MDCs)</b> (lab-scale) 93.3 mg SAC/ g MDC-4h; 99.2 mg SAC/ g MDC-6 h; 94.5 mg SAC/ g MDC-12 h	(Seo et al., 2016) (Song et al., 2018)



Treatment technique	Main mechanism	SAC optimal removal performance	Reference
Chemical		<b>4. UV/TiO<sub>2</sub></b> (lab-scale) UV-light-emitting diode(LED)/TiO <sub>2</sub> has higher SAC removal efficiency and is 16 times more energy efficient than UV-blacklight (BL)/TiO <sub>2</sub>	(Davididou et al., 2018)
		<b>5. UV-A/TiO<sub>2</sub></b> (lab-scale) >90 % degraded and 49% mineralized	(Zelinski et al., 2018)
Fenton Treatment	Organics oxidized and degraded by strong oxidants (usually ·OH) generated by reaction of peroxides (mainly H <sub>2</sub> O <sub>2</sub> ) with ferrous ions (Fe <sup>2+</sup> )	<b>1. Photo/TiO<sub>2</sub>-Fenton</b> (lab-scale) 93 % of TOC removal	(Chen et al., 2014)
		<b>2. Photo-Fenton</b> (lab-scale) 0.05-0.1 L/kJ (reaction rate constant <i>k</i> ,conventional) 2.21-7.88 L/kJ (reaction rate constant <i>k</i> ,assisted by Ethylenediamine-N, N-disuccinic acid) 0.13 L/kJ (reaction rate constant <i>k</i> ,assisted by olive mill wastewater)	(Davididou et al., 2019)
		<b>3. Electro-Fenton</b> (lab-scale) Apparent rate constant <i>k</i> <sub>app</sub> for SAC degradation: 0.18/min (with dimensionally stableanodes (DSA)); 0.19/min (with platinum (Pt) anode); 0.21/min (with boron-doped diamond (BDD) anode) Mineralization for SAC: 55.8 % (with DSA anode); 76.1 % (with Pt anode); 96.2 % (with BDD anode)	(Lin et al., 2016)

Treatment technique	Main mechanism	SAC optimal removal performance	Reference
Biological Bacteria	SAC as sole carbon and energy sources for bacterial aerobic growth	<b>1. Pure strain</b> (lab-scale) Specific growth rate ( $\mu$ ) of 0.14/h of bacterium <i>Sphinogomonas xenophaga</i> SKN under aerobic conditions, and the substrate utilization concomitant with growth.	(Schleheck and Cook, 2003)
	SAC as source for bacterial growth	<b>2. Mixed bacteria</b> (sampling study +lab-scale) Sobo drink containing 0.25g SAC with 50% degradation rate on 12th day; 0.5g SAC with 64.5% degradation rate on 3rd day; 1.0g SAC with 19.3% degradation rate on 12th day; 1.5g SAC with 8.1% degradation rate on 12th day; 2.0g SAC with 3.1% degradation rate on 12th day Bacteria accounting for SAC degradation are common spoilage bacteria and coliform bacteria including <i>Bacillus subtilis</i> , <i>Bacillus pumilis</i> , <i>Bacillus azotomonas</i> , <i>Micrococcus varians</i> , <i>Aeromonas hydrophila</i> , <i>Enterobacter aeromonas</i> , <i>Lactobacillus acidophilus</i> .	(Kabir et al., 2018)
	SAC degraded by nitrifying bacteria together with their induced non-specific oxidative enzymes	<b>3. Nitrifying consortia</b> (lab-scale) 60-80% SAC degraded by nitrifying community	(Tran et al., 2014b)
	SAC as sole carbon source with microbial enzymes initiating SAC degradation	<b>4. Consortia from activated sludge in WWTPs</b> (lab-scale) Complete removal followed zero-order kinetics by consortia dominated by a $\beta$ -proteobacterial genome from the family <i>Rhodocyclaceae</i> .	(Deng et al., 2019)

Treatment technique	Main mechanism	SAC optimal removal performance	Reference
<b>Biological Plant</b>	Highwater solubility and hydrophilic nature of SAC	Uptake by plants (vegetables, i.e. radish, celery, young celery, spinach and cabbagemustard) significant when SAC occurred at high levels in the surrounding environment (sampling study)	(Baalbaki et al., 2017)
<b>Activated sludge</b>	SAC as carbon and energy sources of microorganism with different functions and metabolic ways under aerobic conditions. SAC removal performance depends on diversity of microbial community.	<p><b>1. Aerobic activated sludge in WWTPs</b> (full-scale and/or lab-scale) 90% - nearly 100% of SAC degraded under aerobic conditions in bioreactor</p> <p><b>2. Anaerobic sludge in WWTPs</b> <i>a.</i> &gt;95% of SAC removal in the fermentation processes but no SAC removal in digested sludge (sampling study) <i>b.</i> No SAC degradation under anaerobic digestion (pilot-scale)</p> <p><b>3. Sludge in constructed wetlands</b> (sampling study) 27.1-53.3% by aerobic consortia in constructed wetlands</p>	<p>(Buerge et al., 2009) (Scheurer et al., 2010) (Subedi and Kannan, 2014) (Tran et al., 2015) (Li et al., 2018a)</p> <p>(Buerge et al., 2011)</p> <p>(Phan et al., 2018)</p> <p>(Vymazal and Dvořáková Březinová, 2016)</p>