

1 **Challenges and opportunities for citrus wastewater management and valorisation: a review**

2

3 ¹Caterina Lucia, ¹Vito Armando Laudicina*, ¹Luigi Badalucco, ¹Antonino Galati, ¹Eristanna

4 Palazzolo, ²Michele Torregrossa, ²Gaspare Viviani, ²Santo Fabio Corsino

5

6 ¹Department of Agriculture, Food and Forest Sciences, University of Palermo, Viale delle Scienze,

7 building 4, 90128 Palermo (Italy);

8

9 ²Department of Engineering, University of Palermo, Viale delle Scienze, building 8, 90128 Palermo

10 (Italy);

11

12 *Corresponding author: Vito Armando Laudicina; Department of Agriculture, Food and Forest

13 Sciences, University of Palermo, Viale delle Scienze, building 4, 90128 Palermo (Italy);

14 email vitoarmando.laudicina@unipa.it; tel. +3909123897074

15

16 **doi.org/10.1016/j.jenvman.2022.115924**

17 **ABSTRACT**

18

19 Citrus wastewaters (CWWs) are by-products of the citrus fruit transformation process. Currently,
20 more than 700 million of m³ of CWWs per year are produced worldwide. Until nowadays, the
21 management of CWWs is based on a take-make-use-dispose model. Indeed, after being produced
22 within a citrus processing industry, CWWs are subjected to treatment and then discharged into the
23 environment. Now, the European Union is pushing towards a take-make-use-reuse management
24 model, which suggests to provide for the minimization of residual pollutants simultaneously with
25 their exploitation through a biorefinery concept. Indeed, the recovery of energy nutrients and other
26 value-added products held by CWWs may promote environmental sustainability and close the
27 nutrient cycles in line with the circular bio-economy perspective. Unfortunately, knowledge about
28 the benefits and disadvantages of available technologies for the management and valorisation of
29 CWWs are very fragmentary, thus not providing to the scientific community and stakeholders an
30 appropriate approach. Moreover, available studies focus on a specific treatment/valorisation
31 pathway of CWWs and an overall vision is still missing.

32 This review aims to provide an integrated approach for the sustainable management of CWWs to be
33 proposed to company managers and other stakeholders within the legislative boundaries and in line
34 with the circular bio-economy perspective. To this aim, firstly, a concise analysis of citrus
35 wastewater characteristics and the main current regulations on CWWs are reported and discussed.
36 Then, the main technologies with a general comparison of their pros and cons, and alternative
37 pathways for CWWs utilization are presented and discussed. Finally, a focus was paid to the
38 economic feasibility of the solutions proposed to date relating to the recovery of the CWWs for the
39 production of both value-added compounds and agricultural reuse.

40 Based on literature analysis an integrated approach for a sustainable CWWs management is
41 proposed. Such an approach suggests that after chemicals recovery by biorefinery, wastewaters
42 should be directly used for crop irrigation if allowed by regulations or addressed to treatment plant.

43 The latter way should be preferred when CWWs cannot be directly applied to soil due to lack of
44 concomitance between CWWs production and crop needs. In such a way, treated wastewater should
45 be reused after tertiary treatments for crop irrigation, whereas produced sludges should be
46 undergone to dewatering treatment before being reused as organic amendment to improve soil
47 fertility. Finally, this review invite European institutions and each Member State to promote
48 common and specific legislations to overcome the fragmentation of the regulatory framework
49 regarding CWWs reuse.

50

51 **Keywords**

52 wastewater reuse; intensive wastewater treatment; extensive wastewater treatment; costs and
53 benefits analysis; agricultural reuse; circular economy model

54 **Abbreviations**

- 55 AGS, aerobic granular sludge
- 56 BOD, biochemical oxygen demand
- 57 CAS, conventional activated sludge
- 58 COD, chemical oxygen demand
- 59 CWTs, centralized waste treatment industries
- 60 CWWs, citrus wastewaters
- 61 EO, essential oil
- 62 MBR, membrane bioreactor
- 63 OLR, organic loading rate
- 64 PHA, polyhydroxyalkanoates
- 65 RBCs, rotating biological contactors
- 66 SBMBR, sequencing batch membrane bioreactor
- 67 TFs, tricking filters
- 68 TSS, total suspended solid
- 69 UWWTD, Urban Wastewater Treatment Directive
- 70 VFAs, volatile fatty acids
- 71 WWTP, wastewater treatment plant

72 **1. Introduction**

73 During last decades, the increase of global consumption of natural resources has moved the
74 attention of researchers and stakeholders toward the achievement of a sustainable development
75 concept, which aims to reduce costs on environment, economy and society. In this context, meeting
76 the international regulations and standards, while simultaneously recovering value-added products
77 from waste streams, are becoming increasingly important factors that push forward the
78 implementation of the circular economy model.

79 In this framework, the agro-food industry has embarked on a path against the waste of raw materials
80 and co-products and is moving from a linear model towards a management approach based on the
81 circular economy paradigm (Mak et al., 2020; Yadav et al., 2022). Agro-food wastes have high
82 potential in the form of energy and nutrients recovery (Vaish et al., 2020). This not only would
83 promote the processes sustainability but also facilitate to close environmental nutrient cycles in line
84 with the circular bio-economy perspective (Mak et al., 2020; Vaish et al., 2020).

85 The citrus industry plays an important role in the agro-food industrial sector. Citrus fruits are among
86 the most widespread crops in the world (Schimmenti et al., 2013). They are grown over an area of
87 11.4 million ha. According to the Statistical Bulletin of Food and Agriculture Organization (FAO),
88 global citrus fruits production from 2011 to 2019 increased by 12% thus reaching a production of
89 nearly 144 million Mg (FAO, 2020). The five most important citrus-producing countries in 2019
90 were China, Brazil, India, Mexico and Spain. On average, more than 20% of the total production of
91 citrus fruits is addressed to industrial processing, primarily oriented to juice production and
92 essential oils (EO) extraction (Zema et al., 2019). From the citrus fruits transformation processes
93 two by-products are obtained: a solid/semisolid fraction, constituted by peels and fruit residues, and
94 the citrus wastewaters (CWWs).

95 In the literature, there are several reviews that addressed solutions for the valorisation of the
96 solid/semisolid fraction. These studies indicated that the solid fraction of citrus fruit is generally
97 addressed for disposal or reused as source of relevant bioactive compounds showing a wide range

98 of health effects, thus making it exploitable by pharmaceutical, cosmetic, and food industries (Nieto
99 et al., 2021). Moreover, it might also be directed to dietary fibres (Zema et al., 2019) and fertilizers
100 production, essential oils, phytochemicals and pectin extraction, as well as applied as absorbent
101 material (Calabrò et al., 2016). However, this fraction, being rich in organic matter, could represent
102 a serious environmental issue due to its potential putrescence, i.e. hard to dispose as waste (Tripodo
103 et al., 2004).

104 CWWs are a mixture of fruit constitution water and process water. They are mainly derived from
105 fruit washing, plants, and device cleaning, as well as cooling, essential oil extraction, and peel
106 drying (Zema et al., 2019). Indeed, they may also include effluents derived from the production of
107 citric acid and pectin, citrus molasses, and peel oil depending on treatment plant type (Sharma et al.,
108 2017). CWWs are characterized by low pH and high electrical conductivity. Moreover, they are rich
109 in organic matter and contain nitrogen, phosphorus, some microelements (B, Al, Fe, Zn, Cu) and
110 traces of essential oils. However, due to the different processes contributing to their formation, the
111 chemical properties of CWWs may be extremely variable, even daily. Until nowadays, the
112 management of CWWs is based on a take-make-use-dispose model. Indeed, after being produced
113 within the citrus processing industry, CWWs are subjected to treatment and then discharged into the
114 environment. Previous studies have estimated that the amount of CWWs accounts for
115 approximately 1 to 17 m³ per ton of processed fruits (Calabrò et al., 2018; Di Trapani et al., 2019)
116 depending on the plant's technology for fruit processing. Apart from the large volume to be
117 handled, the management of CWWs represent serious constrains from both an environmental and
118 economic point of view, because of the intrinsic characteristics of such wastewater. Indeed, the low
119 pH (<4.5) (Tamburino et al., 2007), high concentration of organic matter (Corsino et al., 2021a) and
120 essential oils (Calabrò et al., 2016), could produce significant environmental impact if proper
121 disposal practices are not implemented. Complying with the current environmental regulations,
122 which are increasingly stringent, implies the application of novel intensive treatments that generate
123 high costs weighing on the industries budget. On the other hand, simplified treatment systems are

124 admitted only in developing countries, due to the different environmental regulations and socio-
125 economic conditions (Zema et al., 2019).

126 Most of the literature studies concerning the CWWs management mainly focuses on the
127 optimization of the treatments aimed at minimizing the environmental impacts of CWWs disposal.
128 However, the global environmental policies on wastewaters are pushing forward not only to
129 minimize their residual pollution concentrations, but also their exploitation by a biorefinery
130 approach and reuse. In this sense, there is an urgent need to move towards wastewater reuse, as
131 demonstrated by the recent publication of the new regulation (Regulation UE 2020/741) which aims
132 to promote the reuse of both municipal and industrial wastewaters especially in the food sector
133 (Shrivastava et al., 2022). Indeed, food sector plays a significant role in the transition from fossil-
134 based linear economy to sustainable circular bio-economy (Mak et al., 2020). As empathized in a
135 recent study, a strategic approach for wastewater reuse is not only to avoid unnecessary use of
136 higher quality water and encourage the reuse of treated water, but also to encompass the
137 possibilities to recover various value-added resources from wastewater (Shrivastava et al., 2022).

138 Among these, the same wastewater (treated or not), could be used for agricultural purposes, since
139 this requires more than 50% of the total fresh water for human consumption (Mateus et al., 2021).

140 Bearing in mind this, several solutions for a sustainable treatment and valorisation of CWWs have
141 been reported in the literature. Recent studies have demonstrated that, by adopting innovative
142 technologies, CWWs can be used for the extraction of value added compounds and recovery of
143 chemicals (Zema et al., 2019), or just treated while minimizing the impact deriving from their
144 release into the environment (Corsino et al., 2021a; Martín et al., 2010). Indeed, CWWs were
145 successfully exploited for the recovery of polyhydroxyalkanoates (PHA) (Corsino et al., 2022), or
146 agricultural reuse (Ioppolo et al., 2020). However, knowledges about the benefits and disadvantages
147 of these technologies are very fragmentary, thus they are not thoroughly known by the scientific
148 community and stakeholders. Moreover, available studies focus on a specific treatment/valorization
149 pathway of CWWs and an overall vision, who's prospective should be toward an integrated

150 approach for a sustainable CWWs management, is still missing. Indeed, the integration of these
151 conversion processes leads to sub-products, which provides an extra benefit in product recovery and
152 overcomes the restrictions of individual stage (Mak et al., 2020). In addition, considering the recent
153 developments of regulations on wastewater reclamation and the need of the citrus industrial sectors
154 to move towards this direction, a focus on the opportunities and perspectives referring CWWs reuse
155 is required.

156 To fill this gap is important to understand at what stage the research on CWWs is and what are the
157 potential future lines of research to be undertaken for their proper disposal or reuse with a special
158 view to an integrated approach among all these technologies.

159 In light of this, this review aims to provide an integrated approach for the sustainable management
160 of CWWs to be proposed to company managers and other stakeholders within the legislative
161 boundaries and in line with the circular bio-economy perspective. To this aim, firstly, the main
162 current regulations on CWWs disposal and reuse, and a concise analysis of citrus wastewater
163 characteristics are reported and discussed. Then, the main technologies that have been implemented
164 over the previous decades for CWWs treatment are reported with a general comparison of their pros
165 and cons, and focusing on the main concerns related to a sustainable management as well as the
166 opportunities to recover, wherever possible, valuable resources. In addition, alternative pathways
167 for CWWs utilization, such as that for crop irrigation, are presented and discussed. Finally, a focus
168 was paid to the economic feasibility of the solutions proposed to date relating to the recovery of the
169 CWWs for the production of both value-added compounds and agricultural reuse.

170

171 **2. Legislative aspects**

172

173 Citrus industries employ large quantities of water for fruits processing. These operations produce
174 effluents that need proper treatments before their disposal because of the high content of organic
175 matter and other compounds (i.e. surfactants, essential oils, etc.), but also due to low pH and high

176 degree of corrosiveness (Suri et al., 2020). Such properties might cause several environmental
177 impacts if CWWs were improperly disposed, including water and groundwater pollution, toxicity
178 for aquatic organisms, proliferation of vectors for diseases, as well as greenhouse gases emissions
179 during their degradation in soil. For the above reasons, CWWs are subject to strict regulations for
180 their disposal. However, nowadays CWWs are usually directly released into water bodies, treated in
181 intensive or extensive treatment plant, or discharged into the public sewer after proper treatments
182 (Zema et al., 2019), which means that the disposal strategies not always depend on local
183 environmental regulations but sometimes more on specific conditions. In general, water use and
184 wastewater disposal are regulated by national and regional legislations transposed by EU laws. The
185 main EU laws on the environment protection that concern industrial wastewater treatment and reuse
186 are the following: Directive 91/271/EEC (Directive 91/271/EEC, 1991) and Regulation (EU)
187 2020/741 (Regulation EU 2020/741, 2020).

188 The Directive 91/271/EEC (Urban Waste Water Treatment Directive - UWWTD) concerns the
189 urban wastewater collection, processing, and discharge, as well as treatment and discharge of
190 wastewater originating from certain industrial sectors. It aims to protect the environment from
191 possible risks associated to wastewaters. This regulation claims that “*discharges from certain*
192 *industrial sectors of biodegradable industrial wastewater...should be subject to appropriate*
193 *requirements*” and “*should be subjected to special authorization*”. These industrial sectors are listed
194 in the Annex III, in which CWWs can be enumerated in the “*Manufacture of fruit and vegetable*
195 *products*”. The Directive 91/271/EEC also defines the minimal requirements and treatment levels
196 necessary for these wastewaters. More specifically, “*...the discharge of industrial wastewater into*
197 *collecting systems and urban wastewater treatment plants is subject to prior regulations and/or*
198 *specific authorizations by the competent authority or appropriate body*” (article 11), and
199 “*...industrial wastewater entering collecting systems and urban wastewater treatment plants shall*
200 *be subject to such pre-treatment*” (Annex I). Therefore, specific requirements in the
201 regulations/authorizations for CWWs discharges into urban wastewater systems are defined under

202 article 11 and Annex IC. The same directive deals with direct discharges under Art. 13 from
203 biodegradable industrial wastewater (Annex III). More precisely, Article 11 of the UWWTD
204 requires Member States to ensure that competent authorities regulate and give prior authorization
205 for the discharge of industrial wastewater into collecting systems and wastewater treatment plants
206 (WWTPs). Such authorizations must ensure that industrial wastewater entering the collecting
207 systems and/or the treatment plants is pre-treated, when necessary, so that the functioning of the
208 plant and the collecting system is not hindered and, thus, that discharges from the plants do not
209 adversely affect the environment. However, the requirements of Article 11 are relatively general
210 and the specific interpretation of how to meet the requirements of this article are defined separately
211 by each Member State. The UWWTD also aims to control the sludge generated in the treatment
212 operations, and to ensure that it can be safely disposed of and, if possible, used in certain
213 applications (e.g. agriculture). This implies that citrus processing industries should be equipped
214 with a wastewater treatment plant aimed at reducing the pollutants load of the processing effluents
215 to comply the requirements imposed by regulation. However, specific limit values for industrial
216 wastewater are not indicated in the EU Directive, hence they are demanded to national or regional
217 authorities.

218 Since in many countries water is becoming an increasingly rare and precious commodity, a more
219 careful and correct management of this resource is needed for industrial processing to reduce
220 simultaneously the quantity of effluents produced (Klemes, 2012). In this context, a new regulation
221 (Regulation UE 2020/741) was recently published and will apply as from the coming June 2023.
222 This regulation aims to promote the reuse of wastewater and to guarantee that reclaimed water is
223 safe for agricultural irrigation, thereby ensuring a high level of protection of the environment and of
224 human and animal health, thus promoting the circular economy, supporting adaptation to climate
225 change. However, this Regulation “...*should not concern biodegradable industrial wastewater from*
226 *plants belonging to the industrial sectors listed in Annex III to Directive 91/271/EEC, unless the*
227 *wastewater from those plants enters a collecting system and is subject to treatment in an urban*

228 wastewater treatment plant". This likely means that industrial wastewater reuse shall be still
229 delegated to local regulations. Generally, these define the intended and the eligible uses for
230 wastewater reuse, including agricultural reuse for irrigation of crops (human and animal
231 consumption) and irrigation of green areas, civil reuse (for washing streets, powering heating or
232 cooling systems, supply of dual supply networks, with the exclusion of the direct use, etc.) and
233 industrial reuse (fire-fighting, process, washing water, etc.). The reclaimed water quality is divided
234 into four classes, for each of which the permitted uses and irrigation methods are set out. Therefore,
235 a crop belonging to a specific category shall be irrigated with reclaimed water of the corresponding
236 minimum reclaimed water quality class, unless additional requirements (article 5) are requested.
237 This results in achieving the quality requirements foreseen in Table 2, which reports the limits for
238 some quality parameters, like *E. coli*, biochemical oxygen demand, total suspended solid (TSS),
239 turbidity, according to the specific water quality class. The quality requirements for wastewater
240 reuse are very stringent. Indeed, one of the crucial issues in the reuse of treated wastewater for crop
241 irrigation is the residual presence of pathogenic microorganisms which represent a potential health
242 risk to consumers when entering in the food chain (Libutti et al., 2018). Regarding microbiological
243 contamination levels, the corresponding guidelines allow unrestricted crop irrigation with a
244 bacteriological effluent quality characterized by less than 10 CFU 100 ml⁻¹ of *E. coli* in 80% of
245 samples. To comply these levels, advanced treatment units (such as filtration, membranes, activated
246 carbon systems, disinfection) are necessary. This entails that wastewater reuse basically depends on
247 the economic viability of the regeneration versus the purchase, use, treatment and discharge of fresh
248 water (Zema et al., 2019).

249

250 **3. Characteristics of CWWs**

251

252 The production of CWWs is characterized by a large quantitative and qualitative variability
253 depending on many factors, such as water consumption per weight unit of processed fruits, amount

254 of citrus processed and overall water management in the plant. This variability can, in turn, change
255 annually because of agricultural production, market trends and plant operations.

256 The yearly total volume of CWWs is affected by the amount of citrus fruits produced during the
257 growing season, which shows both inter- and intra-annual variability (Zema et al., 2019). In the
258 Mediterranean areas, more than 70% of citrus fruit is processed during the trimester February-April,
259 with highest peaks usually set on March (Figure 1) due to the wide production of citrus fruits
260 (Corsino et al., 2018; Zema et al., 2019). The quantitative variability of CWWs changes also weekly
261 and daily because of the plant downtimes during night and weekend. Recently, Zema et al. (2019)
262 estimated a yearly production of more than 700 million of m³ of CWWs from 2017 due to the
263 increase of citrus fruit intended for the industry. Corsino et al. (2018) reported for an Italian citrus
264 factory processing 25 tons h⁻¹ of citrus fruits (lemons, oranges and tangerines), a production of
265 about 17 m³ of CWWs per tons of processed fruits. Rosas-Mendoza et al. (2018) have estimated in
266 the northern part of the State of Veracruz (Mexico, USA) a production of CWWs in the range from
267 0.79 to 1.25 m³ for each tons of oranges processed. However, more data collected (Rosas-Mendoza
268 et al., 2018) have shown values of 1.58–3 m³ of CWWs produced for each tons of fruit processed.

269 This wide range of water consumption in citrus industries depends on several factors mainly related
270 to the technologies for fruit processing implemented in the factory, the number of processing lines
271 operating simultaneously, processing technologies (e.g., juice, EO, or pulp extraction) and type of
272 fruit (Bozzano et al., 2021). In general, recirculation of water for specific operations (e.g., cooling,
273 EO extraction, etc.) allows saving large volume of fresh water, thus minimizing that of CWWs
274 production. For instance, the cooling towers with open-cycle system requires large quantity of
275 freshwater, thus generating significant volume of CWWs. In contrast, water-cooling closed system
276 allow to significantly reduce wastewater production (Zema et al., 2018).

277 Concerning the qualitative variability, it depends not only on the type and stage of fruit ripeness but
278 on the technological and construction characteristics of the transformation plant too. The main
279 parameters measured for CWWs characterization are (Table 1) pH, biochemical and chemical

280 oxygen demand (BOD and COD, respectively), total nitrogen (TN), total phosphorus (TP), TSS,
281 soluble potassium (K), magnesium (Mg) and calcium (Ca), essential oils (EOs) (Corsino et al.,
282 2018; Koppa and Pullammanappallil, 2013a). Generally, washing, and cooling operations produce
283 wastewater with a low pollutants load, whereas other processing units (e.g., EO, fruit extraction)
284 generate a significant increase in the organic pollutant's concentration. Cleaning and cooling
285 operations (e.g., machineries and equipment's) produce a dilution effect referring to some
286 parameters (BOD, COD, EO) while increasing, in some cases, the concentration of others
287 (surfactants, pH, TP, etc.) depending on the type of chemicals used. Values of COD and TSS are
288 very variable depending on the various stages of the transformation processes (Corsino et al., 2022).
289 Data available in the literature referring to COD concentration indicate a wide range of variation
290 between approximately 5000-75000 mg L⁻¹. This could depend on the sampling point, since CWWs
291 generated from juice and EO extraction, or peel dehydration are characterized by very high organic
292 matter concentration (>60-70 g L⁻¹) (Garcia et al., 2019), but also on the plant process design. In
293 general, recirculation of water in fruit processing units generate CWWs with higher pollutants
294 concentration, whereas water open-cycle systems determine a significant dilution. Minor variations
295 could be due to the type of fruit processed.

296 CWWs have acid reaction due to the high content of acidic compounds among which the most
297 abundant are citric and malic acids (Sharma et al., 2017). Variations in pH of CWWs could be
298 partly attributable to the type of fruit (Zema et al., 2019), but more in general to the use of alkaline
299 chemicals for cleaning operations. Based on the operating volume of the equalization unit,
300 wastewater generated from cleaning operations could buffer the pH of CWWs. Similarly, the
301 amount of TN and TP are very variable, although their concentrations in relation to that of COD are
302 very low (C/N >1000). It is worth to observe that CWWs are characterized by the presence of other
303 trace element in not negligible concentrations. The presence of such elements makes CWWs
304 interesting for agricultural purposes to improve soil fertility (Ioppolo et al., 2020).

305

306 **4. Potential use and reuse of CWWs**

307

308 Citrus fruit wastes consist of solid/semisolid fraction and CWWs. The latter can be i) exploited for
309 the recovery of chemicals through biorefinery (Fazzino et al., 2021; Sharma et al., 2017), ii)
310 addressed to lagooning (Zema et al., 2019), iii) addressed to wastewater treatment plant (Corsino et
311 al., 2018; Martín et al., 2010), or iv) reused in agriculture for crop irrigation (Ioppolo et al., 2020)
312 (Figure 2). Techniques and methods for the recovery of chemicals are well established, studies
313 about aerobic and anaerobic CWWs treatment are still in progress, whereas those about lagooning
314 and their reuse in agriculture are still at the beginning. Chemicals recovered from CWWs are
315 phenolic acids, flavonoids, carotenoids, limonoids and essential oils. Such chemicals are extracted
316 from CWWs by using solvents with different polarity, followed by precipitation or centrifugation
317 (Sharma et al., 2017).

318

319 4.1 CWWs storage

320 The variability of the raw CWWs characteristics could represent a critical issue for the WWTP
321 since they would operate under variable conditions that might result in poor purification
322 performances especially during the load peaks and transition periods (start-up and end of citrus
323 season). The reason for this is that some processes (e.g., biologic units, secondary clarifiers) might
324 not allow excessive hydraulic or organic loads. Therefore, WWTPs serving citrus processing
325 industries should be equipped with proportioning and equalization units to limit and minimize the
326 effects of load variations. Equalization is a method of retaining wastewater in a basin so that the
327 effluent discharged is fairly uniform in its quality characteristics, whereas proportioning means the
328 discharge of CWW in proportion to the flow treatment capacity of the WWTP (Nemerow, 2007). In
329 most of cases, it is possible to combine equalization and proportioning in the same basin. A
330 secondary but significant effect is that of lowering the concentration of effluent pollutants,
331 especially if during cyclic productions of citrus industries large quantity of fresh water is used (e.g.,

332 floor and equipment washing). In addition, as pH varies along the day, since usually alkaline
333 chemicals are used at the end of the production cycle, an efficient equalization unit could
334 considerably reduce the needs of chemicals used for pH neutralization. Indeed, CWWs produced
335 during the fruit processing phases are characterized by low pH (3-4.5), whereas once the fruit
336 processing has finished the cleaning operations produce wastewater characterized by very high pH
337 (>11). Thus, the buffering capacity of the latter wastewater could result in a substantial cost saving
338 for chemicals supply.

339 Nonetheless, storage of not treated CWWs is necessary also when the purpose is their direct reuse
340 for crop irrigation. Indeed, the production of CWWs may span a long period, from days to months,
341 and not totally coincide with crop water needs. The storage of CWWs requires the existence of
342 facilities and suitable places (Bonari et al., 2007). During the storage, CWWs may undergo to
343 fermentation process due to the high content of carbohydrates and scarcity of oxygen. Fermentation
344 led to the production of gases such as bio-hydrogen (Zema et al., 2019) and methane (Calabrò et al.,
345 2016), and also volatile fatty acids as result of hydrolysis, acidogenesis and acetogenesis processes
346 (Corsino et al., 2021a). To prevent the occurrence of biological activities in the storage units,
347 fermentation inhibitors could be used. Efficient inhibitors may be furfural, 5-
348 hydroxymethylfurfural, acetic, lactic or formic acids (Narendranath et al., 2001). However, if
349 fermentation inhibitors are used, the quality of CWWs has to be evaluated before their further uses.

350

351 4.2 Intensive CWW treatments

352

353 According to EU regulations (Directive 91/271/EEC, 1991) industrial wastewaters could be
354 discharged into urban collecting systems after appropriate preliminary treatments. The most
355 developed techniques for industrial wastewaters purification are intensive treatments carried out in
356 WWTP located within the production site. The principle with these processes is to intensify the
357 phenomena of the transformation and dissimilation of organic matter as occur naturally, thereby

358 allowing to operate on a reduced surface area (Guo et al., 2016). Intensive treatments could be
359 performed in different ways by means of several technologies. The most used intensive technology
360 in WWTPs for CWWs treatment is the conventional activated sludge (CAS) process, although other
361 intensive technologies such as rotating biological contactors (RBCs) or tricking filters (TFs) are
362 available (Matamoros et al., 2016). In some cases, especially for small potential WWTPs, extended
363 aeration system, which is a modification of the CAS process with a higher sludge retention time, is
364 adopted. More rarely, advanced treatments are applied (i.e., MBR, membrane bioreactor) and only
365 for special requirements (i.e., wastewater reuse, small space availability, etc.). Intensive biological
366 treatments are characterized by several drawbacks mainly linked to the variability of CWWs
367 characteristics and the presence of toxic compounds (e.g., EO), the long-start up times necessary to
368 achieve steady-state conditions when the plant is reactivated after the not operating period
369 (summer), and the high energy requirements ($> 2 \text{ kWh kgCOD}^{-1}$). More details about the
370 applications and comparison of the main biological processes are provided in the following
371 sections. Alternative treatment systems to biologic processes are based on physical and/or chemical
372 (i.e., clarification-flocculation, concentration by evaporation, advanced oxidation processes). These
373 are of limited use for CWWs, since these plants are expensive and have higher energy requirements
374 than biologic based processes (Zema et al., 2019). Therefore, biological processes are still the most
375 used for CWWs treatment.

376 Another possibility for CWWs handling is the disposal in centralized waste treatment industries
377 (CWTs). The CWTs handle both wastewater treatment residuals and industrial process by-products
378 that come from several industries. CWT facilities receive a wide variety of hazardous and non-
379 hazardous industrial wastes for treatment. CWWs treatment with other effluents could be a viable
380 option to reduce the drawbacks related to their treatment in a dedicated WWTP. Indeed, by
381 blending CWWs with municipal and/or other industrial wastewater it could be possible to reduce
382 the concentration of toxic compounds, overcome the issue related to the seasonal variability and

383 exploit synergistic effects deriving from the co-treatment with other effluents (e.g, blending with
384 wastewater with low C/N ratio).
385 CWWs treatment at a WWTP located within the production site implies both high capital costs for
386 all the facilities construction (200-700 € PE⁻¹), as well as operating ones for all the management
387 activities necessary for CWW treatment (2.0 kWh kg COD_{removed}⁻¹). In contrast, treatment of CWWs
388 in CWTs entails high disposal (100-300 € m⁻³) and transportation costs, whose incidence on the
389 overall industry economy depends on the distance from the disposal site. Consequently, the choice
390 between these two alternatives is based mainly on economic criteria.

391 All these treatment processes involve several challenges, including high-energy consumption,
392 financial costs and environmental impact, as well as a need to be resilient to a periodic variability of
393 the wastewater characteristics. Alternative management/treatment approaches for some of the above
394 processes could help to meet these challenges by reducing the economic and environmental burden
395 as well as turning wastewater into valuable resource. Accordingly, the traditional approach whereby
396 wastewater must be purified sufficiently to meet the environmental regulations, is going to be
397 moved to the concept of biorefinery, in which waste streams are used as a valuable substrate with
398 concomitant water treatment. The implementation of the biorefinery concept could change the
399 open-model of industrial activity towards a more virtuous path in which those components of
400 wastewaters, which have value, could be reused. The biorefinery concept is a driving force towards
401 the implementation of cleaner production in the industrial sector.

402 In the following sections, a comprehensive review of the main technological solutions for CWWs
403 treatment and valorization is provided in view of a sustainable, synergic and integrated approach
404 consistently with the biorefinery and circular economy models.

405

406 4.2.1 CWWs anaerobic treatment

407

408 Anaerobic treatment is one of the most common methods for agro-food wastes management
409 (Sharma et al., 2017). In fact, this process makes possible the simultaneous removal of the organic
410 pollution, the reduction of waste streams to be disposed and the production of high-value added by-
411 products (Rosas-Mendoza et al., 2020). The biorefinery approach involving CWWs might have a
412 high potential impact on the development of circular economy in the citrus-processing sector.
413 Because of the high organic matter content, CWWs have an attractive bioenergy and bio-products
414 potential. Indeed, several studies have focused on volatile fatty acids, hydrogen, and methane
415 production from CWWs, through the combination of dark fermentation and anaerobic digestion
416 processes (Lukitawesa et al., 2018; Torquato et al., 2017). However, due to the presence of
417 inhibiting compounds, such as essential oils (D-limonene), the application of anaerobic treatments
418 aimed at the valorisation of CWWs for energy purposes is challenging (Calabrò et al., 2020; Zema
419 et al., 2019). Therefore, specific pre-treatments, such as hydrothermal treatment (Saadatinavaz et
420 al., 2021), or dilution with other liquid streams, like fruits and machineries cleaning waters (Corsino
421 et al., 2021a), are generally performed before anaerobic treatment to decrease the concentration of
422 such inhibiting compounds. Alternatively, given the high-market value of such products, an
423 integrated biorefinery approach could be advisable to recover essential oils before performing
424 anaerobic treatments. Indeed, EO has promising potential application in several sectors from the
425 food industry (for production of pectin, dietary fibres, etc.), to the cosmetic and pharmaceutic
426 Industries (extraction of flavonoids, flavouring agents and citric acid) (Zema et al., 2018). However,
427 in many cases, these uses are still not economically sustainable. Several studies have focused on the
428 development of new separation techniques for the chemical, food and pharmaceutical industries and
429 lately received a lot of attention due to the increasing energy prices and the drive to reduce CO₂
430 emissions. In this sense, a process based on microwave hydro-diffusion permits fast and efficient
431 extraction, reduces waste, avoids water and solvent consumption, and allows substantial energy
432 savings (Bousbia et al., 2009).

433 Anaerobic digestion is defined as the biological conversion under reducing conditions of organic
434 matter into a variety of products, including biogas. Biogas is a mixture of gases mainly composed
435 by CH₄ (40–65%) and CO₂ (35–60%) along with other minor components (H₂O, H₂S, NH₃) (Rosas-
436 Mendoza et al., 2018). Among these, methane is the most attractive compound within biogas since
437 its potential use for renewable energy. Nevertheless, an accurate control of the main operating
438 parameters is necessary during the digestion process, as a function of the composition of CWWs
439 and the aims of the process. A down-flow stationary fixed film anaerobic digester was successfully
440 operated for 76 days under thermophilic (55°C) conditions. At an average organic loading rate
441 (OLR) of 0.51 kg COD/m³ d⁻¹ and a hydraulic retention time of 16 days, the reactor yielded 2.1
442 Nm³ of biogas per m³ of treated wastewater. No long term toxicity issues due to limonene were
443 observed (Koppar and Pullammanappallil, 2013a). A recent study tested a high-rate anaerobic
444 hybrid reactor to perform the anaerobic digestion of effluents from a citrus industry (Rosas-
445 Mendoza et al., 2018). The reactor obtained high soluble and total COD removal (85%), as well as
446 high methane yields close to 0.15 L CH₄ g COD_{removed}⁻¹, operating at an OLR of 8 g COD L⁻¹d⁻¹.
447 Nevertheless, the authors observed an inhibitory effect on methanogenic bacteria due to the
448 presence of D-limonene since no pre-treatment was performed for elimination of this essential oil.
449 This caused a decrease in the COD removal and methane yields, especially when the reactor was
450 operated at high OLR. However, the authors stressed that the configuration of the reactor allows
451 handling high organic loads, and, due to the presence of the biofilm, the inhibitory effect of D-
452 limonene can be minimized.

453 To minimize the effect of D-limonene on methane production, a two-stage anaerobic digestion with
454 internal recirculation was proposed (Lukitawesa et al., 2018). The effluent from the first stage was
455 filtered to separate and discharge the solid phase rich in D-limonene from the liquid one containing
456 less D-limonene, that was fed in the second stage. A higher methane yield (160–203 NmL gVS⁻¹,
457 where VS are volatile solids) was observed compared with a control reactor without the solid-phase
458 separation (60–133 NmL gVS⁻¹). The effect of D-limonene removal through hydrothermal process

459 was recently investigated (Saadatinavaz et al., 2021). The bio-methane produced from the untreated
460 orange waste (OW) residue was 194 NmL gVS⁻¹, higher than that from the pre-treated residues. The
461 reason might be the removal of hemicellulosic sugars and other biodegradable materials when the
462 organic waste was subjected to pre-treatment and enzymatic hydrolysis. In another study, the
463 removal of D-limonene through a solvent-extraction method produced a significant increase of the
464 methane yield (Battista et al., 2020). The authors demonstrated that the methane potential of
465 extracted and unextracted orange peels was comparable (355–365 NL CH₄ kgVS⁻¹), although the
466 orange peels without a previous limonene extraction took twice as long to reach the final methane
467 production. All the studies seem to confirm that removal of essential oils prior to anaerobic
468 digestion is crucial for maximize bio-methane productivity and kinetics. Nevertheless, these
469 treatments should not involve other compounds, especially biodegradable materials. Improvement
470 of process kinetics is of a matter of importance especially in order to reduce the plants footprint.
471 Higher process kinetic would require smaller reactors or the possibility of increasing the treatment
472 capacity of existing ones.

473 Volatile fatty acids (VFAs) are one of the largely used compounds in the chemical industry that
474 serve as starting materials for biofuel production and for the synthesis of a variety of products, such
475 as biopolymers, reduced chemicals, and derivatives (Strazzera et al., 2018). VFAs have a wider
476 range of applications, from the food, pharmaceutical, and cosmetics industries to biofuels (e.g.,
477 biobutanol) and bioplastics (polyhydroxyalkanoates) production (Hunter et al., 2021).

478 VFA production from CWWs was studied in MBR operating at OLR up to 8 g VS L⁻¹d⁻¹
479 (Lukitawesa et al., 2021). VFAs were mainly constituted by acetate, whereas the fraction of
480 butyrate, caproate, and propionate was lower. Without performing any pre-treatment, the highest
481 yield of VFAs, 0.67 gVFA g VS⁻¹, was achieved at OLR 4 g VS L⁻¹d⁻¹. Contrarily, when CWW was
482 pre-treated to remove D-limonene, the VFA yield increased to VFAs 0.84 gVFA gVS⁻¹ operating at
483 the same OLR. At higher OLR (8 g VS L⁻¹d⁻¹) the authors observed a sharp decrease in yield only
484 for the untreated CWW. Moreover, Corsino and co-authors evaluated the effect of operating

485 conditions on VFA production from CWW subjected to dilution with other processing effluents
486 (Corsino et al., 2021a). The authors found that acetate production was maximized by operating
487 under unbalanced nutrients (C: N: P = 200:0.1:0.1), without removing the particulate fraction and
488 operating at pH higher than 5. The authors stressed that dilution with other processing streams
489 enabled to minimize the effects of the essential oils on process yield and kinetics. Similarly, in
490 another study it was observed the VFA production from orange peel fermentation after limonene
491 recovery was close to 0.35 g VFA gTS⁻¹ after 5 days only (Battista et al., 2020), where TS are total
492 solids. Among the other by-products achievable from anaerobic treatments, the hydrogen (H₂) may
493 be a valuable resource to be used for local energy supply, reducing the operational costs of citrus
494 industries facilities. The bioconversion of CWWs to hydrogen through dark fermentation was
495 recently reported in the literature (Torquato et al., 2017). The authors demonstrated that CWWs
496 showed significantly higher potential for H₂ production when compared to synthetic and domestic
497 wastewaters, resulting in a H₂ bioconversion efficiency close to 73%. In another study, the
498 hydrogen production from CWWs was maximized by applying an electroporation treatment at
499 different intensity levels (30-120 kWh m⁻³) to achieve methanogen inactivation (Karim et al., 2019).
500 In comparison with other pre-treatments, the highest hydrogen production of 896 mL was achieved
501 with the electroporation treatment, followed by sonication with a probe (678 mL), sonication in a
502 bath (563 mL) and heat-shock treatment (545 mL).

503 Overall, anaerobic treatments could be considered a preliminary stage for CWWs since the residual
504 organic pollution is not suitable for their release into the environment. Thus, additional downstream
505 treatments are required to meet the standards imposed by the environmental regulations.

506 Nonetheless, it is worth to stress that anaerobic treatment of high-strength wastewater allows not
507 only to reduce the overall energy requirement necessary to comply the environmental regulations
508 but additionally it provides synergistic effects deriving from the integration with other treatments
509 (e.g., biologic aerobic processes). For instance, volatile fatty acids represents the ideal substrate for

510 the enrichment of mixed microbial culture with microorganisms able to produce PHA in aerobic
511 downstream systems (Argiz et al., 2020).

512

513 4.2.2 CWWs aerobic treatment

514

515 Aerobic biological treatments are generally considered as an alternative to the anaerobic ones for
516 CWWs treatment.

517 Aerobic biological treatments are generally based on conventional activated sludge (CAS) systems
518 or on biofilm processes (i.e. trickling filters). Although these processes are widely applied for the
519 treatment of municipal wastewater, their replication for CWWs treatment is challenging (Zema et
520 al., 2019). Indeed, specific characteristics of CWWs makes conventional processes unsuitable to
521 meet the discharge limits imposed by European regulations (Directive 91/271/EEC, 1991). The
522 main drawbacks of aerobic processes referring to CWWs treatment are related to the high organic
523 matter content, nutrients imbalance, seasonal and weekly flow rate variability, high energy
524 requirements especially for aeration system, etc. (Di Trapani et al., 2019). Moreover, some specific
525 characteristics of CWWs, such as the high availability of readily biodegradable organic matter, low
526 pH and imbalance of carbon/nitrogen ratio are favourable for the occurrence of dysfunction in the
527 CAS process (Jenkins et al., 2003). Filamentous and viscous bulking, as well as biological foaming
528 are high risk factors for a CAS based treatment of CWWs. These dysfunctions could occur
529 simultaneously and could create severe issues in the solid-liquid separation phase (Corsino et al.,
530 2018).

531 Nutrient imbalance could be easily addressed by adding nitrogen (N) and phosphorous (P) to
532 achieve a COD:N:P ratio close to 200:5:1 (Metcalf and Eddy, 2015). This could prevent the
533 occurrence of viscous bulking (Wanner, 2017). Filamentous bulking and biological foaming are
534 more difficult to be addressed since many of the trigger factors are peculiarities of CWWs. In such

535 cases, application of metabolic selection methods (i.e. anoxic or anaerobic selectors) could prevent
536 the overgrowth of specific filamentous organisms (i.e. *Nocardia Amarae*-Like Organisms, NALO).
537 Seasonal and daily variability is a considerable drawback for biological based processes. Indeed, it
538 could cause significant load variations, occurrence of process dysfunctions, affect the microbial
539 community structure and consequently the treatment's efficiency. A well-designed storage and
540 equalization unit is necessary to prevent load-shock during daily and weekly flow fluctuation. Large
541 equalization basins could reduce the CWWs variability and limit the instability and breakdown of
542 the processes. This could also be helpful to exploit the buffering capacity of some liquid streams
543 deriving from routinely operation within the production process (i.e. a storage tank washing with
544 sodium hydroxide), thereby reducing the amount of chemicals necessary for pH neutralization
545 before the biological treatment (Hawash et al., 1988). Nevertheless, because of the high space
546 requirement, their integration within the production site is not always feasible. In addition, seasonal
547 variation of CWWs quantities produces drawbacks during two transit times corresponding to the
548 increase or decrease of CWWs production: i) during the plant start-up (winter period) and ii) at the
549 end of citrus season. In the first cases, the sudden increase of the food to microorganism ratio could
550 cause the onset of dispersed growth (Wanner, 2017), since the high availability of organic substrate
551 decrease the production of extracellular polymeric substances by bacteria, reducing the activated
552 flocs size. On the other hand, in the second case, the long starvation period (low F/M and long
553 sludge retention time) could cause the onset of pinpoint floc, which consists of the formation of
554 small flocs ($< 50 \mu\text{m}$) with poor settling properties. The long-term loading changes caused larger
555 disturbances to the floc size distribution than more rapid but shorter ones (Barbusiński and
556 Kościelniak, 1995). Generally, after the substrate overload occurred, the flocs are more prone to
557 breakup, thus increasing the effluent turbidity. In such cases, the addition of flocculating agent
558 could be a temporary solution to mitigate the problem. A modular WWTP constituting by different
559 biologic reactors operating in parallel could be a practical solution to have a flexible system able to
560 be adapted to load variations. In addition, depending on the storage capacity available and on the

561 plant design, CWWs with high organic content (e.g., peel drying or EO extraction) could be stored
562 during the winter and gradually treated in the WWTP during the summertime. Moreover, it could be
563 suggested to reduce the aeration rate or switching off on briefly times a day to reduce bacterial
564 endogenous respiration and limit sludge stabilization. Certainly, this practice loss its effectiveness
565 as the standstill time increases (Edwards and Norman, 2015). Other techniques, such as the addition
566 of enzymatic products or activated sludge from other plants, could be used to speed up the start-up
567 phase, although these are very expensive and of limited applicability (Folino et al., 2018).

568 The high-energy requirements for aerobic treatment of CWWs still remain a serious drawback. The
569 energy requirement of these plants can be close to $2.0 \text{ kWh kg COD}^{-1}$ ($0.86 \text{ kgCO}_2 \text{ kgCOD}^{-1}$), that
570 is about four times greater than that required for municipal wastewater treatment (Zema et al.,
571 2019), mainly due to the high concentration of readily biodegradable organic matter in soluble
572 form. The integration of anaerobic with aerobic treatments in a synergistic way could help to
573 minimize the overall impact of intensive treatments, while maximizing the energy and material
574 recovery from CWWs treatment. In this sense, the integration with anaerobic upstream treatment
575 could noticeably reduce the energy requirement for aeration since the most of organic matter is
576 removed without oxygen requirement and more it allows recovering energy through biogas
577 production. Indeed, anaerobic treatment could reduce the organic content of CWWs for the
578 downstream aerobic process, while producing biogas suitable for energetic purposes (e.g., heat or
579 electric energy production) and effluent enriched in volatile fatty acids. The effluent from AD could
580 be treated in intensive aerobic processes with a double advantage: i) handling with effluent enriched
581 in VFA could be beneficial to enrich the activated sludge with PHA-storing bacteria, thus offering a
582 further pathway for excess sludge valorization (PHA recovery), and ii) decreasing of the organic
583 content reduces the energy requirements.

584 Another approach studied to minimize the environmental impact of CWWs aerobic treatment is the
585 implementation of advanced processes. Advanced aerobic technologies such as aerobic granular
586 sludge (AGS) and MBR were successfully adopted for CWWs treatment both in single and

587 sequential stage. Corsino et al. (2018) examined the treatment of CWWs in two AGS reactors
588 operating at OLR ranging between 3.0-15 kg COD m⁻³d⁻¹ and at pH of 7.0 and 5.5. The authors
589 found that high COD removal (> 90%) could be achieved by operating at OLR not higher than 7 kg
590 COD m⁻³d⁻¹ and pH close to neutral conditions, while ensuring an appropriate balance between the
591 feast and famine phases to prevent the overgrowth of fast-growing microorganisms (e.g.,
592 filamentous bacteria) and ensure the granules stability in the long term. AGS allows reducing
593 significantly the energy costs for wastewater treatment being close to 0.35 kWh kgCOD⁻¹ (0.15
594 kgCO₂ kgCOD⁻¹) (Giesen et al., 2013). AGS was also coupled with MBR technology in a in series
595 AGS/MBR configuration (Di Trapani et al., 2019). This scheme was also compared to the
596 conventional MBR one. The results demonstrated that both plant configurations enabled very high
597 COD removal, with average values close to 99%. Nevertheless, higher fouling tendency was
598 observed in the AGS/MBR configuration due to AGS deflocculation. This might severely affect the
599 membrane service life. A recent study evaluated the treatment of CWWs in a sequencing batch
600 membrane bioreactor (SBMBR) with activated sludge enriched in microorganisms able to store
601 intracellular biopolymer such as polyhydroxyalkanoate (PHA) (Corsino-Membranes). The authors
602 demonstrated that the enrichment of the sludge with PHA-storing bacteria was favoured by the
603 characteristics of CWWs (high soluble COD availability, low nitrogen content) that enabled to
604 mitigate the fouling behaviour of the membrane and to achieve high removal performances at high
605 OLR (> 3 kgCOD m⁻³d⁻¹). Among the available studies, MBR allowed obtaining the highest COD
606 removal from CWWs. The specific treatment costs of MBR technology are close to 0.7-1.0 kWh
607 kgCOD⁻¹ (0.30-0.43 kgCO₂ kgCOD⁻¹). However, the lack of knowledge about the use of MBR for
608 CWWs treatment in large-scale application requires additional studies aimed at evaluating the
609 hydraulic performances of the membrane in the long-term, the need of chemicals for fouling
610 maintenance and the overall management costs. Nevertheless, considering the high-quality of the
611 effluent from a MBR, the application of this technology should be advisable only for the purpose of

612 reuse of CWWs. Wastewater reuse within the same industry or for agricultural purposes could
613 significantly contribute to save fresh water supporting a more sustainable use of water resource.

614

615 4.2.3 Sludge production

616 The excess sludge production is another drawback of intensive biological treatments of CWWs.
617 Indeed, the excess sludge generated from the transformation of the organic matter into new biomass
618 entails high expenditure for its treatment and disposal. The handling of excess sludge in municipal
619 wastewater treatment plant is a considerable economic burden, accounting for 30–40% of the total
620 capital cost and 50% of plant operation costs (Valentino et al., 2015). Sludge production from
621 CWWs treatment through aerobic process was estimated about 0.10-0.30 kg VS kg COD⁻¹ (Corsino
622 et al., 2018). Considering that the average COD concentration in CWWs is between 5-27 kg COD
623 m⁻³ (Zema et al., 2018), the specific productivity of excess sludge could range between 0.5-9.0 kg
624 SS m⁻³ of treated wastewater. This entails that a considerable amount of excess sludge must be
625 treated and disposed, thereby affecting the overall operating costs related to CWWs treatment. To
626 reduce the impact of sludge treatment on the overall operating costs, innovative solutions aimed at
627 reducing the excess sludge production or valorizing the sludge once produced should be considered
628 (Collivignarelli et al., 2019). The use of efficient sludge dewatering and drying system might
629 considerably decrease the amount of sludge to be disposed. In this context, thermal drying systems
630 of excess sludge allow evaporating significant amount of water in the sludge thereby reducing the
631 final weight to be disposed (Zhu et al., 2022). In the frame of citrus industries, there are several
632 thermal waste streams that could be used as a heat source (e.g. waters from boilers or heaters). This
633 could reduce the need of heat to carry out thermal drying of sludge, thereby minimizing the use of
634 conventional fossil fuels. Considering the high organic content of the sludge produced during
635 CWWs treatment, energy recovery through biogas production in anaerobic digester or agricultural
636 utilization after composting could be a feasible solution in line with the circular bio-economy
637 model. In this sense, sludge could be used as a fertilizer since the organic nitrogen and phosphorous

638 in bio-solids are used quite efficiently by crops upon the mineralization process. Moreover, the
639 supply of organic matter is one of the most important agro-technique to improve structure,
640 minimize erosion, increase water holding capacity and counteract fertility decline of soils of the
641 semiarid Mediterranean environment (Laudicina et al., 2012; Palazzolo et al., 2019).

642

643 3.4 Extensive treatment

644

645 Extensive treatments of wastewaters have both advantages and disadvantages. Indeed, if on the one
646 hand, these are attractive solutions considering the lower construction costs, energy requirements
647 and reliable purification efficiency, on the other hand, the need of wide areas, the long hydraulic
648 retention time and, possible unpleasant smells due to anaerobic processes are the main constraints
649 related to their use (Carawan et al., 1979; Kimball, 1999). Among the extensive treatments,
650 lagooning is the most widely used (Andiloro et al., 2021; Koppa and Pullammanappallil, 2013a;
651 Lobato et al., 2013). This system is used for the treatment of several types of wastewaters, including
652 agricultural and industrial ones (Kruzic and Liehr, 2008). Indeed, it has proved to be a valid
653 alternative to the treatment of CWWs with positive results (Andiloro et al., 2013).

654 A lagoon can be defined as a greater or lesser deep basin in which wastewaters are stored.

655 Lagooning refers to a low-cost and efficient treatment process, which requires lower management
656 and mechanical equipment (Andiloro et al., 2021; Zema et al., 2019). It is based on the wastewaters
657 self-purification by aerobic and/or anaerobic microorganisms activity (Andiloro et al., 2021; Lobato
658 et al., 2013).

659 CWWs can be treated in anaerobic or aerobic biological ponds. The anaerobic treatment is
660 recommended far away from private homes due to the possible production of unpleasant smells and
661 long times and volumes of pond. In order to face issues related to anaerobic treatment, the aerobic
662 one is generally adopted (Indelicato et al., 1997; Kimball, 1999).

663 However, according to Zema et al. (2019) the best option for CWWs treatment is the use of aerobic-
664 anaerobic aerated lagoon systems, which allow higher purification efficiency and lower energy
665 demand ($0.57 \text{ kWh kgCOD}^{-1} - 0.24 \text{ kgCO}_2 \text{ kgCOD}^{-1}$) when the following measures are adopted: i)
666 high depth of pond, ii) optimization of operating hours according to the oxygen demand, and iii)
667 aeration only during the night when the cost of energy is cheaper. Although lagooning can represent
668 a suitable natural solution for the treatment of CWWs (Zema et al., 2016), water is lost thus not
669 allowing its reuse.

670 Among the environmental factors affecting extensive treatments, the temperature is one of the main
671 having a significant effect on depuration performances. Basically, increase in the temperature of the
672 wastewater caused a change in solubility of oxygen in water (decrease in the saturation
673 concentration C_s), acceleration of the process of the oxygen adsorption, the activity rate of bacteria
674 and the rate of gases which are transferred to and from water (Alisawi, 2020). In general, the higher
675 removal performances in extensive treatment systems could be obtainable during the warm periods
676 (Kadlec and Reddy, 2001). Zema et al. investigated the effect of temperature in aerated ponds
677 (Zema et al., 2012) and observed that the differences between for the removal rate of COD during
678 the autumn-winter period compared to summer period were significant, so confirming the
679 significant influence of temperature. Because the maximum production of CWWs is during the
680 colder months, performances of such systems could be affected by low temperature. Nevertheless,
681 the greater effect of temperature in extensive treatments is on the nutrients removal (nitrogen and
682 phosphorous) (Alisawi, 2020), thus considering the absence of such elements in CWWs, the effect
683 of temperature is of lesser importance.

684 In addition to the temperature, even precipitations play a crucial role in the operation of extensive
685 treatment. A previous study observed that rainfall improved final effluent water quality of an
686 aerobic lagoon system, although this was shown to be through dilution rather than improvement of
687 treatment efficacy. Specifically, following precipitation events the contaminant removals were

688 negatively impacted in the aerobic lagoon, whereas the removal rates were increased for anaerobic
689 condition with the rainfall dilution (Alisawi, 2020).

690

691 3.5 Agricultural reuse of CWWs

692

693 Water is a critical input for agricultural production and plays an important role in food security.

694 Irrigated agriculture represents 20 percent of the total cultivated land and contributes 40 percent of

695 the total food produced worldwide (Ashley and Gruère, 2021). Due to population growth,

696 urbanization, and climate change, the competition for water resources is expected to increase, with a

697 particular impact on agriculture. Moreover, at the same time, the demand of water for the

698 agricultural production to assure food for the increasing world population is expected to increase

699 (El-Zanfaly, 2015).

700 The agricultural sector has proved to be the most suitable for the use of wastewater. Indeed, the

701 application of wastewater for crop irrigation has grown a lot in recent years reaching about 20

702 million ha of irrigated land worldwide (Khalid et al., 2018).

703 According to Barbagallo et al. (2012) the water demand for irrigation is not satisfied properly in

704 some areas of the Mediterranean basin due to the increase of adverse conditions such as drought,

705 degradation of water body quality and ever-growing citizen demand.

706 Several factors contribute to define the correct use of wastewater. Wastewater originated from

707 industrial, commercial, domestic, and dairy farm show different composition and variability in

708 quantitative and quality aspects. This may have implications on the wastewater impact on the soil,

709 which depends on soil characteristics, plant species and nature of wastewaters (El-Zanfaly, 2015).

710 Thus, to use these effluents for agricultural practice, it is necessary to have knowledge of climatic,

711 pedological and cultural conditions of a specific territory as well as wastewater characteristics

712 (Bonari et al., 2007). Holding nutrients essential for plant growth such as nitrogen, phosphorus and

713 potassium, as well as soluble organic matter (Table 1), their use, as they are, for crop irrigation

714 could enhance soil fertility and crop production. Such an aspect is of paramount importance as on
715 the one hand it allows to close the nutrients cycle and, on the other hand, to add organic matter to
716 soil. Moreover, the use of not treated CWWs in agriculture for crop irrigation may be a valid
717 alternative to their treatment in plant (Zema et al., 2019), if allowed by regulations. However,
718 studies focusing on the effects of CWWs on soil fertility and crop growth are very few.

719 Recently, Ioppolo et al. (2020) investigated the effects of not treated CWWs on soil chemical
720 properties and microbial community at laboratory scale. Lemon, orange and tangerine wastewaters,
721 diluted with water at the rate of 1/3 or 2/3 or as they were (3/3), were applied once to soil samples
722 to bring them at 50% of the water holding capacity. Soils were analysed at three different times
723 during the 56 days of incubation since CWWs addition. Soil reaction decreased from 2 to 3 pH units
724 following the addition of CWWs but, after 7 days, it recovered reaching values of the control
725 (distilled water). The authors attributed such a reduction to the high content of organic acids in
726 CWWs. At the same time, also electrical conductivity showed a transient increase and, therefore,
727 the Authors suggested the need of monitoring the electrical conductivity (EC) if CWWs are added
728 repeatedly.

729 Total and labile organic C increased following the addition of CWWs proportionally to the amount
730 stored in the different CWWs (Ioppolo et al., 2020). Such an increase was, as for pH and EC,
731 ephemeral although after 56 days of incubation soil moistened with not diluted orange wastewater
732 had more organic C than the control. Total and extractable organic C increased following the
733 addition of CWWs proportionally to the amount stored in the different CWWs. The increase of total
734 and labile organic C following the addition of CWWs, in turn, stimulated microbial biomass and
735 activity (CO₂ emission) although in ephemeral way (Ioppolo et al., 2020). Indeed, soil
736 microorganisms live generally in oligotrophic conditions, thus when fresh organic substrates are
737 added, such as those holding in CWWs, they restart their activity (Laudicina et al., 2012, 2013).

738 Based on the above results, Ioppolo et al. (2020) suggested a possible role of CWWs in sustainable
739 farming. However, such a possibility has to be properly evaluated considering repeated addition of

740 CWWs, CWWs production time vs. crop needs, organic matter and minerals addition, and different
741 type of soil and crops.

742

743 **4. CWWs: costs and benefits**

744

745 The reuse of wastewater lies within the principle of the circular economy model, which provides the
746 recovery of resources considered as waste, achieving at the same time decrease of waste disposal
747 and develop of value-added products (Corsino et al., 2021a).

748 Financial advantages occur in wastewater reuse throughout correct agronomic and processing
749 operations aimed at the exploitation of wastewater for supporting natural water sources and
750 reducing fertilizer costs, because of higher content in nutrients and the possibility to cultivate
751 multiple crops with the greater water productivity (El-Zanfaly, 2015). The high content of nutrients
752 in the wastewaters makes them particularly suitable for the irrigation of crops such as citrus and
753 olives in Gaza district, resulting in a 70% reduction of fertilization costs and an increase in profits
754 (Nassar, 2008). A recent study performed in Morocco assessed the economic feasibility of the
755 irrigation and nutrition of citrus plants by using treated wastewaters (Oubelkacem et al., 2020).

756 Cost-effectiveness of agronomic use of wastewater depends on the transfer phase from the
757 processing industry to land application. Indeed, if CWWs are carried in small-medium distances
758 and soils are in proper conditions, the agricultural utilization of this water could be cost-effective
759 solution (Zema et al., 2012). Moreover, results show that the main obstacle to the use of treated
760 wastewaters is linked to their higher cost, due to the treatment process, compared to fresh water
761 (0.23 € m^{-3} vs 0.15 € m^{-3}). Thus, a reduction in the price could encourage their use on the 59% of
762 the cultivated area. In terms of costs, a significant element is represented by the type of treatment
763 performed on the wastewater. Relating to treatment of 4 million cubic meters (mcm)/year, the
764 activated sludge treatment cost is about $\text{€ } 0.18 \text{ m}^{-3}$ (per annum), with an addition of $\text{€ } 0.12 \text{ m}^{-3}$
765 annually for nitrification-denitrification stage (Haruvy, 1997). In particular, as emphasized by

766 Navarro et al. (2008), evaluating the feasibility of a pilot plant for the treatment of wastewater from
767 the lemon processing industry, the treatment cost is mainly related to the energy cost used for
768 optimal oxygen transfer in the plant (70% of the operating costs). In light of this, Guzmán et al.
769 (2016) found that the adoption of a photo-Fenton powered by renewable energy sources (solar) is a
770 promising techniques for the treatment of CWW with a cost of 13.8 € m⁻³, which include both the
771 cost of operation and amortization, lower than that detected for conventional treatment processes.
772 With a view to optimizing processes and reducing the volume of water arising from the citrus fruit
773 processing process, Shen et al. (2021), by improving segment membrane removal process, obtain a
774 processing water rich in electrolytes, pectin and flavonoids usable as drinkable beverages at low
775 costs. As the authors themselves point out, although the cost of the new membrane is higher than
776 the conventional one, the system allows for production of new electrolyte beverages with high
777 health benefits and profitable on the market. It is reasonable thinking that the lower the treatment
778 cost, the greater the interest in their use. However, this concept clashes with possible negative
779 effects on the environment and in particular on groundwater pollution. As emphasized by Haruvy,
780 (2005) the treatment cost of wastewater reflects the environmental costs linked to the groundwater
781 pollution. Whatever the case, if it is decided to deal with wastewater, it is desirable to conduct an
782 analysis of costs, risks, and benefits (Haruvy, 1997). In addition to the economic dimension,
783 unquestionably important for the potential reuse of CWWs in agriculture, the environmental
784 dimension associated with the recovery of wastewater, currently considered a waste, and the
785 nutrients held which can improve soil fertility must be considered. The reuse of treated or not
786 treated CWWs in agriculture represents, indeed, a strategic solution in line with the circular
787 bioeconomy paradigm where a waste product derived from material of biological origin becomes a
788 resource by improving eco-efficiency, reducing the demand for fertilizers, and enhancing the waste
789 stream.

790

791 4.1 Advantages of CWWs reuse

792

793 The reuse of CWWs shows environmental, agronomic, and economic advantages. In arid and
794 semiarid environment, agricultural production is limited by the scarcity of water or by its
795 unsuitability for crop irrigation due to high concentration of soluble salts (Ungureanu et al., 2020).
796 Thus, the use of wastewaters may represents a compelling necessity (Barbagallo et al., 2012). Using
797 wastewaters for irrigation could avoid an increasingly massive subtraction of natural sources of
798 water from the environment, thus allowing its full use for civil and industrial purposes (Barbagallo
799 et al., 2012). Furthermore, the use of wastewaters agrees with the principles of the circular economy
800 model in both solid and liquid wastes management practices because it provides the simultaneous
801 minimization of waste disposal and generation of value-added products (Lee et al., 2014).
802 From the agronomic point of view, the reuse of CWWs is advantageous because allows to supply, at
803 the same time, water, organic matter and mineral plant nutrients (Laudicina et al., 2013). CWWs are
804 rich of low molecular weight organic substrates readily available for soil microorganisms. Indeed,
805 recently, Ioppolo et al. (2020) demonstrated that the addition of lemon, orange and tangerine
806 wastewaters stimulated soil microbial biomass and activity even when they were diluted before to
807 be applied. Such an aspect is of paramount importance because it improves the nutrient cycling
808 (Laudicina et al., 2012). On the other hand, the great amount of carbohydrates they hold may work
809 as an organic cement, thus improving soil aggregation and enhancing soil fertility (Palazzolo et al.,
810 2019; Ren et al., 2022). Among the main mineral plant nutrients supplied by CWWs, all nitrogen,
811 phosphorus and potassium are the most abundant. Thus, when CWWs are used for crops irrigation,
812 supply of mineral fertilizers can be rescaled to take into account the amount of mineral nutrients
813 added by CWWs.

814 From an economic point of view, the reuse of CWWs can be advantageous, both for the citrus
815 processing industry and farms. The former, supplying CWWs to farmers, can save the cost for their
816 treatment and displacement, the latter, instead, can reduce their operating cost not paying money to

817 buy water. In addition to the above advantages, since CWWs are available at the soil surface,
818 farmers may reduce the cost to pump the volume of water needed for irrigation practice (Jaramillo
819 and Restrepo, 2017). Finally, the application of CWWs on soil provides nutrients reducing
820 inorganic fertilizers rates required for plants growth. The amount of nutrients that can be reached,
821 considering a release of wastewater approximately of 5,000 m³ ha⁻¹ year, is almost 250 kg ha⁻¹ for
822 N, 50 kg ha⁻¹ for P and 150 kg ha⁻¹ for K. Moreover, CWWs can provide some micronutrients such
823 as B, Fe, Al, Zn, Cu (Becerra-Castro et al., 2015). These CWW characteristics inevitably contribute
824 to reducing the operating costs related to the purchase of fertilizers.

825

826 4.2 Disadvantages of CWWs reuse

827

828 As for advantages, also disadvantages can be environmental, agronomic and economic.

829 Environmental disadvantages of CWWs reuse are limited to the possible higher mobility of heavy
830 metals due to their high acidity. Indeed, organic acids held by CWWs may increase heavy metals
831 mobility by forming chelates (Violante et al., 2010) or by speeding up silicates alteration, thus
832 making free aluminium hydroxides (Qin et al., 2018).

833 The decrease of the pH and the concomitant increase in electrical conductivity (Ioppolo et al., 2020)
834 are the two main disadvantages from an agronomic point of view because both limit the range of
835 cultivable agricultural crops. However, if soil has a good buffer capacity, small changes or rapidly
836 recover of pH is expected (Ioppolo et al., 2020), thus doing such disadvantage transient.

837 According to Leverenz et al. (2011), however, the major constraints to reuse CWWs are noticed in
838 (i) the long distance between the treatment facility and the agricultural lands, (ii) the construction
839 costs of the pipe system for water displacement and, (iii) the necessary storage of CWWs during
840 winter season, considering that they are produced in a period when there is not a high demand of
841 water for crop growth (Leverenz et al., 2011). Other constraints may be (i) higher costs of treatment
842 processes and reclaimed wastewater; (ii) protection of environment and human health; (iii)

843 inadequate regulations to reuse of wastewater; (iv) higher costs for personnel and monitoring
844 equipment; (v) lack of proper cooperation between authorities on the treatment and reuse of
845 wastewater; (vi) distrust of farmers and consumers on this practice (El-Zanfaly, 2015).

846 Moreover, fragmentation of the regulatory environment in the field of CWW reclamation could
847 limit its spread across Europe. Removing this fragmentary approach might provide better
848 improvement of public perception and raising confidence for wastewater reuse.

849

850 **5. Concluding remarks and future perspectives**

851

852 The industrial processing involving citrus fruits generates high amount of CWWs that have to be
853 properly disposed. This literature review analysis highlights the numerous opportunities associated
854 with the use of CWWs but also the most common obstacles. The latter related in a specific way to
855 the high disposal costs, suggesting the necessity to find alternatives that allow companies to make a
856 profit or, otherwise, to reduce costs, without neglecting the possible environmental implications of
857 wastewater use. The alternatives to disposal may be the reuse of CWWs for agricultural purpose,
858 after their exploitation for chemicals recover. The advantages of such alternative far outweigh the
859 disadvantages because, firstly, water is completely recovered and, secondly, organic matter and
860 mineral nutrients are added to soil thus improving soil fertility and allowing the closure of the
861 nutrient cycle. In such a way, the reuse of CWWs for agricultural purpose fits with the new
862 guidelines of the European Union about the circular bioeconomy, allowing the transition from the
863 take-make-use-dispose model to the take-make-use-reuse one. On the other side, CWWs subject to
864 treatment processes can give rise to high value by-products, which can compensate high costs
865 necessary for their treatment. Despite these potentialities, to date, very few studies evaluated the
866 economic feasibility of the use of CWWs in agriculture, addressed to understand their impact on the
867 management costs of companies, or the opportunities related to the production of high value-added
868 compounds.

869 In light of this review, future research should focus on parameters set up during the aerobic
870 digestion for reducing sludge production, the effect of repeated addition of CWWs on soil chemical
871 and biochemical properties, and crop yield, with a particular attention to the dynamics of heavy
872 metals. In addition, economic and environmental feasibility analyses of the potential solutions for
873 the use of CWWs are necessary to encourage green investments in the sector from a circular bio-
874 economy perspective. From this point of view, and in the case of the use of CWWs for agricultural
875 purposes, it could be interesting to estimate the market value of nutrients and organic matter added
876 to the soil possibly replacing those that the farmer would have had to use, and the impact on the
877 reduction of operating costs for this replacement. Furthermore, with reference to the treatment of
878 CWWs to obtain valuable by-products, it is essential to enrich the empirical literature of studies on
879 the economic analysis of transformation or treatment processes, in order to provide entrepreneurs
880 with useful information and solid data that may justify their investments.

881 Based on above considerations, the best environmental and economic way to process CWWs could
882 be that reported in Figure 3. Thus, after chemicals recovery by biorefinery, wastewaters should be
883 directly used for crop irrigation if allowed by regulations or addressed to treatment plant. The latter
884 way should be preferred when CWWs cannot be directly applied to soil due to lack of concomitance
885 between CWWs production and crop needs. In such a way, treated wastewater should be reused
886 after tertiary treatments for crop irrigation, whereas sludges should be undergone to dewatering
887 treatment before being reused as organic amendment to improve soil fertility.

888 Finally, conclusions emerging from this review invite European institutions and each Member State
889 to promote common and specific legislations to overcome the fragmentation of the regulatory
890 framework regarding CWW reuse.

891

892 **References**

893

894 Alisawi, H.A.O., 2020. Performance of wastewater treatment during variable temperature. Appl.

895 Water Sci. 10, 1–6. doi:10.1007/s13201-020-1171-x

896 Andiloro, S., Bombino, G., Denisi, P., Folino, A., Zema, D.A., Zimbone, S.M., 2021. Depuration
897 performance of aerated tanks simulating lagoons to treat olive oil mill wastewater under
898 different airflow rates, and concentrations of polyphenols and nitrogen. *Environ. - MDPI* 8 (8),
899 70. doi:10.3390/environments8080070

900 Andiloro, S., Bombino, G., Tamburino, V., Zema, D.A., Zimbone, S.M., 2013. Aerated lagooning
901 of agro-industrial wastewater: depuration performance and energy requirements. *J. Agric. Eng.*
902 44, 0–5. doi:10.4081/jae.2013.s2.e166

903 Argiz, L., Fra-Vázquez, A., del Río, Á.V., Mosquera-Corral, A., 2020. Optimization of an enriched
904 mixed culture to increase PHA accumulation using industrial saline complex wastewater as a
905 substrate. *Chemosphere* 247. doi:10.1016/j.chemosphere.2020.125873

906 Ashley, C., Gruère, G., 2021. 7th Roundtable on Financing Agricultural Water, Sustainable use of
907 water for agriculture - Co-convened with FAO 27-28 January 2021 1–18.

908 Aziz, T.N.A.T., Tajuddin, R.M., Kamarun, D., Kordi, N.E., Malini, R., 2018. Citrus fruit waste
909 leachate treatment by using newly developed flat sheet membrane. *AIP Conf. Proc.* 2020.
910 doi:10.1063/1.5062671

911 Barbagallo, S., Cirelli, G.L., Consoli, S., Licciardello, F., Marzo, A., Toscano, A., 2012. Analysis
912 of treated wastewater reuse potential for irrigation in Sicily. *Water Sci. Technol.* 65, 2024–
913 2033. doi:10.2166/wst.2012.102

914 Barbusiński, K., Kościelniak, H., 1995. Influence of substrate loading intensity on floc size in
915 activated sludge process. *Water Res.* 29, 1703–1710. doi:10.1016/0043-1354(94)00326-3

916 Battista, F., Remelli, G., Zanzoni, S., Bolzonella, D., 2020. Valorization of Residual Orange Peels:
917 Limonene Recovery, Volatile Fatty Acids, and Biogas Production. *ACS Sustain. Chem. Eng.*
918 8, 6834–6843. doi:10.1021/acssuschemeng.0c01735

919 Becerra-Castro, C., Lopes, A.R., Vaz-Moreira, I., Silva, E.F., Manaia, C.M., Nunes, O.C., 2015.
920 Wastewater reuse in irrigation: A microbiological perspective on implications in soil fertility

921 and human and environmental health. *Environ. Int.* 75, 117–135.
922 doi:10.1016/j.envint.2014.11.001

923 Bonari, E., Ercoli, L., Silvestri, N., Carcea, G., Barresi, F., 2007. Linee guida per l'utilizzazione
924 agronomica delle acque di vegetazione e delle acque reflue da aziende agroalimentari. Agenzia
925 per la protezione dell'ambiente e per i servizi tecnici (APAT), Rome, Manuali e linee guida
926 ISBN: 978-88-448-0301-8.

927 Bousbia, N., Vian, M.A., Ferhat, M.A., Meklati, B.Y., Chemat, F., 2009. A new process for
928 extraction of essential oil from Citrus peels: Microwave hydrodiffusion and gravity. *J. Food*
929 *Eng.* 90, 409–413. doi:10.1016/j.jfoodeng.2008.06.034

930 Bozzano, G., Raymo, M., Manenti, F., Rulli, M.C., Giroto, F., Piazza, L., 2021. Prompting
931 sustainability in the citrus derivatives industry: A case study. *Clean. Eng. Technol.* 4, 100127.
932 doi:10.1016/j.clet.2021.100127

933 Calabrò, P.S., Fazzino, F., Sidari, R., Zema, D.A., 2020. Optimization of orange peel waste ensiling
934 for sustainable anaerobic digestion. *Renew. Energy* 154, 849–862.
935 doi:10.1016/j.renene.2020.03.047

936 Calabrò, P.S., Pontoni, L., Porqueddu, I., Greco, R., Pirozzi, F., Malpei, F., 2016. Effect of the
937 concentration of essential oil on orange peel waste biomethanization: Preliminary batch results.
938 *Waste Manag.* 48, 440–447. doi:10.1016/j.wasman.2015.10.032

939 Calabrò, P.S., Pontoni, L., Porqueddu, I., Greco, R., Pirozzi, F., Malpei, F., Rigby, H., Smith, S.R.,
940 TeKippe, R.J., Thevendraraj, S., Klemeš, J., Paz, D., Aso, G., Cardenas, G.J., Rosas-
941 Mendoza, E.S., Méndez-Contreras, J.M., Martínez-Sibaja, A., Vallejo-Cantú, N.A., Alvarado-
942 Lassman, A., Corsino, S.F., Di Trapani, D., Torregrossa, M., Viviani, G., Akrotos, C.S.,
943 Tekerlekopoulou, A.G., Vayenas, D. V., Zema, D.A., Fòlino, A., Zappia, G., Calabrò, P.S.,
944 Tamburino, V., Zimbone, S.M., Koppa, A., Pullammanappallil, P., Martín, M.A., Siles, J.A.,
945 Chica, A.F., Martín, A., Ruiz, B., Flotats, X., Zema, D.A., Andiloro, S., Bombino, G.,
946 Tamburino, V., Sidari, R., Caridi, A., Fagbohunge, M.O., Herbert, B.M.J., Hurst, L., Li, H.,

947 Usmani, S.Q., Semple, K.T., 2018. Agro-industrial wastewater treatment with decentralized
948 biological treatment methods. *Waste Manag.* 33, 8993–8999.
949 doi:10.1016/j.biortech.2016.04.106

950 Carawan, R., Chambers, V., Zall, R.R., Wi, R.H., 1979. Fruit and Vegetable Water and
951 Management. Annual Report n. AM-18E.

952 Collivignarelli, M.C., Abbà, A., Miino, M.C., Torretta, V., 2019. What advanced treatments can be
953 used to minimize the production of sewage sludge in WWTPs? *Appl. Sci.* 9, 2650.
954 doi:10.3390/app9132650

955 Corsino, S.F., Di Trapani, D., Capodici, M., Torregrossa, M., Viviani, G., 2021a. Optimization of
956 acetate production from citrus wastewater fermentation. *Water Resour. Ind.* 25, 100140.
957 doi:10.1016/j.wri.2021.100140

958 Corsino, S.F., Di Trapani, D., Torregrossa, M., Viviani, G., 2018. Aerobic granular sludge treating
959 high strength citrus wastewater: Analysis of pH and organic loading rate effect on kinetics,
960 performance and stability. *J. Environ. Manage.* 214, 23–35.
961 doi:10.1016/j.jenvman.2018.02.087

962 Corsino, S.F., Di Trapani, D., Torregrossa, N., Piazzese, D., 2021b. Preliminary evaluation of
963 biopolymers production by mixed microbial culture from citrus wastewater in a MBR system
964 using respirometric techniques. *J. Water Process Eng.* 41, 102003.
965 doi:10.1016/j.jwpe.2021.102003

966 Corsino, S.F., Di Trapani, D., Traina, F., Cruciatà, I., Scirè Calabrisotto, L., Lopresti, F., La
967 Carrubba, V., Quatrini, P., Torregrossa, M., Viviani, G., 2022. Integrated production of
968 biopolymers with industrial wastewater treatment: Effects of OLR on process yields,
969 biopolymers characteristics and mixed microbial community enrichment. *J. Water Process
970 Eng.* 47, 102772. doi:10.1016/j.jwpe.2022.102772

971 Di Trapani, D., Corsino, S.F., Torregrossa, M., Viviani, G., 2019. Treatment of high strength
972 industrial wastewater with membrane bioreactors for water reuse: Effect of pre-treatment with

973 aerobic granular sludge on system performance and fouling tendency. *J. Water Process Eng.*
974 31, 100859. doi:10.1016/j.jwpe.2019.100859

975 Directive 91/271/EEC, 1991. The urban waste water treatment directive. Available online on
976 <https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A31991L0271>.

977 Edwards, D.C., Norman, P.E., 2015. Best practices for management of wastewater treatment
978 operations during extended refinery shutdowns, in: 88th Annual Water Environment
979 Federation Technical Exhibition and Conference, WEFTEC 2015.
980 doi:10.2175/193864715819539966

981 El-Zanfaly, H.T., 2015. Wastewater reuse in agriculture: a way to develop the economies of arid
982 regions of the developing countries. *J. Environ. Prot. Sustain. Dev.* 1, 144–158.

983 FAO, 2020. Citrus Fruit Fresh and Processed Statistical Bulletin. doi:10.5860/choice.36-2167

984 Fazzino, F., Mauriello, F., Paone, E., Sidari, R., Calabrò, P.S., 2021. Integral valorization of orange
985 peel waste through optimized ensiling: Lactic acid and bioethanol production. *Chemosphere*
986 271. doi:10.1016/j.chemosphere.2021.129602

987 Folino, A., Tamburino, V., Zappia, G., Zema, D.A., Zimbone, S.M., 2018. Valorisation of Citrus
988 Processing Waste : a Review. *Waste Manag.* 80, 252–273.

989 Garcia, C.F.H., de Souza, R.B., de Souza, C.P., Fontanetti, C.S., 2019. Effluent from Citrus
990 Industry: Toxic Parameters of Orange Vinasse. *Water. Air. Soil Pollut.* 230.
991 doi:10.1007/s11270-019-4260-4

992 Giesen, A., van Loosdrecht, M., de Bruin, B., van der Roest, H., Pronk, M., 2013. Full-scale
993 Experiences with Aerobic Granular Biomass Technology for Treatment of Urban and
994 Industrial Wastewater, in: International Water Week.

995 Guzmán, J., Mosteo, R., Sarasa, J., Alba, J.A., Ovelleiro, J.L., 2016. Evaluation of solar photo-
996 Fenton and ozone based processes as citrus wastewater pre-treatments. *Sep. Purif. Technol.*
997 164, 155–162. doi:10.1016/j.seppur.2016.03.025

998 Haruvy, N., 2005. Land Use and Water Management in Israel- Economic and Environmental

999 Analysis of Sustainable Reuse of Wastewater in Agriculture. 45th Congr. Eur. Reg. Sci. Assoc.
1000 L. Use Water Manag. Sustain. Netw. Soc. 23-27 August 2005.

1001 Haruvy, N., 1997. Agricultural reuse of wastewater: Nation-wide cost-benefit analysis. *Agric.*
1002 *Ecosyst. Environ.* 66, 113–119. doi:10.1016/S0167-8809(97)00046-7

1003 Hawash, S., Hafez, A.J., El - Diwani, G., 1988. Citrus Processing Wastewater Treatment. *Chemie*
1004 *Ing. Tech.* 60, 128–130. doi:10.1002/cite.330600214

1005 Hunter, S.M., Blanco, E., Borrion, A., 2021. Expanding the anaerobic digestion map: A review of
1006 intermediates in the digestion of food waste. *Sci. Total Environ.* 767, 144265.
1007 doi:10.1016/j.scitotenv.2020.144265

1008 Indelicato, M., Tamburino, V., Zimbone, S.M., 1997. Prove di invaso ed irrigazione con acque
1009 reflue dell'industria agrumaria, in: Proceedings of "Convegno Nazionale Di Ingegneria
1010 Agraria. Ancona 11-12 September.

1011 Ioppolo, A., Laudicina, V.A., Badalucco, L., Saiano, F., Palazzolo, E., 2020. Wastewaters from
1012 citrus processing industry as natural biostimulants for soil microbial community. *J. Environ.*
1013 *Manage.* 273, 111137. doi:10.1016/j.jenvman.2020.111137

1014 Jaramillo, M.F., Restrepo, I., 2017. Wastewater reuse in agriculture: A review about its limitations
1015 and benefits. *Sustain.* 9 (10), 1734. doi:10.3390/su9101734

1016 Jenkins, D., Richard, M.G., Daigger, G.T., 2003. Manual on the Causes and Control of Activated
1017 Sludge Bulking, Foaming and Other Solids Separation Problems. IWA, London ISBN
1018 1566706475.

1019 Kadlec, R.H., Reddy, K.R., 2001. Temperature Effects in Treatment Wetlands. *Water Environ. Res.*
1020 73, 543–557. doi:10.2175/106143001x139614

1021 Karim, A., Islam, M.A., Mohammad Faizal, C.K., Yousuf, A., Howarth, M., Dubey, B.N., Cheng,
1022 C.K., Rahman Khan, M.M., 2019. Enhanced Biohydrogen Production from Citrus Wastewater
1023 Using Anaerobic Sludge Pretreated by an Electroporation Technique. *Ind. Eng. Chem. Res.* 58,
1024 573–580. doi:10.1021/acs.iecr.8b03586

1025 Khalid, S., Shahid, M., Natasha, Bibi, I., Sarwar, T., Shah, A.H., Niazi, N.K., 2018. A review of
1026 environmental contamination and health risk assessment of wastewater use for crop irrigation
1027 with a focus on low and high-income countries. *Int. J. Environ. Res. Public Health* 15, 1–36.
1028 doi:10.3390/ijerph15050895

1029 Killham, K., 1994. *Soil ecology*. Cambridge University Press. doi:10.1017/9780511623363

1030 Kimball, D., 1999. *Citrus Processing :A Complete Guide*, 2nd ed, Materials Research Bulletin.

1031 Klemes, J.J., 2012. Industrial water recycle/reuse. *Curr. Opin. Chem. Eng.* 1, 238–245.
1032 doi:10.1016/j.coche.2012.03.010

1033 Koppar, A., Pullammanappallil, P., 2013a. Anaerobic digestion of peel waste and wastewater for on
1034 site energy generation in a citrus processing facility. *Energy* 60, 62–68.
1035 doi:10.1016/j.energy.2013.08.007

1036 Koppar, A., Pullammanappallil, P., 2013b. Anaerobic digestion of peel waste and wastewater for on
1037 site energy generation in a citrus processing facility. *Energy* 60, 62–68.
1038 doi:10.1016/j.energy.2013.08.007

1039 Kruzic, A.P., Liehr, S.K., 2008. Natural Treatment and Onsite Systems. *Water Environ. Res.* 80,
1040 1206–1224. doi:10.2175/106143008x328608

1041 Laudicina, V., Palazzolo, E., Badalucco, L., 2013. Natural Organic Compounds in Soil Solution:
1042 Potential Role as Soil Quality Indicators. *Curr. Org. Chem.* 17, 2991–2997.
1043 doi:10.2174/13852728113179990120

1044 Laudicina, V.A., Barbera, V., Gristina, L., Badalucco, L., 2012. Management practices to preserve
1045 soil organic matter in semiarid mediterranean environment, in: *Soil Organic Matter: Ecology,*
1046 *Environmental Impact and Management*. New York : Nova Science Publishers, Inc., pp. 39–
1047 61.

1048 Lee, W.S., Chua, A.S.M., Yeoh, H.K., Ngoh, G.C., 2014. A review of the production and
1049 applications of waste-derived volatile fatty acids. *Chem. Eng. J.* 235, 83–99.
1050 doi:10.1016/j.cej.2013.09.002

- 1051 Leverenz, H.L., Tchobanoglous, G., Asano, T., 2011. Direct potable reuse: A future imperative. *J.*
1052 *Water Reuse Desalin.* 1, 2–10. doi:10.2166/wrd.2011.000
- 1053 Libutti, A., Gatta, G., Gagliardi, A., Vergine, P., Pollice, A., Beneduce, L., Disciglio, G., Tarantino,
1054 E., 2018. Agro-industrial wastewater reuse for irrigation of a vegetable crop succession under
1055 Mediterranean conditions. *Agric. Water Manag.* 196, 1–14. doi:10.1016/j.agwat.2017.10.015
- 1056 Lobato, J., González del Campo, A., Fernández, F.J., Cañizares, P., Rodrigo, M.A., 2013.
1057 Lagooning microbial fuel cells: A first approach by coupling electricity-producing
1058 microorganisms and algae. *Appl. Energy* 110, 220–226. doi:10.1016/j.apenergy.2013.04.010
- 1059 Lukitawesa, Eryildiz, B., Mahboubi, A., Millati, R., Taherzadeh, M.J., 2021. Semi-continuous
1060 production of volatile fatty acids from citrus waste using membrane bioreactors. *Innov. Food*
1061 *Sci. Emerg. Technol.* 67, 102545. doi:10.1016/j.ifset.2020.102545
- 1062 Lukitawesa, Wikandari, R., Millati, R., Taherzadeh, M.J., Niklasson, C., 2018. Effect of effluent
1063 recirculation on biogas production using two-stage anaerobic digestion of citrus waste.
1064 *Molecules* 23, 1–11. doi:10.3390/molecules23123380
- 1065 Mak, T.M.W., Xiong, X., Tsang, D.C.W., Yu, I.K.M., Poon, C.S., 2020. Sustainable food waste
1066 management towards circular bioeconomy: Policy review, limitations and opportunities.
1067 *Bioresour. Technol.* 297, 122497. doi:10.1016/j.biortech.2019.122497
- 1068 Martín, M.A., Siles, J.A., Chica, A.F., Martín, A., 2010. Biomethanization of orange peel waste.
1069 *Bioresour. Technol.* 101, 8993–8999. doi:10.1016/j.biortech.2010.06.133
- 1070 Matamoros, V., Rodríguez, Y., Albaigés, J., 2016. A comparative assessment of intensive and
1071 extensive wastewater treatment technologies for removing emerging contaminants in small
1072 communities. *Water Res.* 88, 777–785. doi:10.1016/j.watres.2015.10.058
- 1073 Mateus, A., Torres, J., Marimon-Bolivar, W., Pulgarín, L., 2021. Implementation of magnetic
1074 bentonite in food industry wastewater treatment for reuse in agricultural irrigation. *Water*
1075 *Resour. Ind.* 26. doi:10.1016/j.wri.2021.100154
- 1076 Metcalf, Eddy, 2015. *Wastewater Engineering Treatment and Resource Recovery*. 5th edition.

1077 ISBN 9780073401188 / 0073401188.

1078 Molinos-Senante, M., Hernández-Sancho, F., Sala-Garrido, R., Cirelli, G., Guo, J., Fu, X., Andrés
1079 Baquero, G., Sobhani, R., Nolasco, D.A., Rosso, D., 2016. Trade-off between carbon emission
1080 and effluent quality of activated sludge processes under seasonal variations of wastewater
1081 temperature and mean cell retention time - European Union Commission, Science of the Total
1082 Environment.

1083 Narendranath, N. V., Thomas, K.C., Ingledew, W.M., 2001. Effects of acetic acid and lactic acid on
1084 the growth of *Saccharomyces cerevisiae* in a minimal medium. *J. Ind. Microbiol. Biotechnol.*
1085 26, 171–177. doi:10.1038/sj.jim.7000090

1086 Nassar, A.G., 2008. Effect of Citrus by-Products Flour Incorporation on Chemical , Rheological
1087 and Organolepic Characteristics of Biscuits. *Sci. Technol.* 4, 612–616.

1088 Navarro, A.R., Lopez, Z.O., Maldonado, M.C., 2008. A pilot plant for the treatment of lemon
1089 industry wastewater. *Clean Technol. Environ. Policy* 10, 371–375. doi:10.1007/s10098-008-
1090 0152-9

1091 Nemerow, N., 2007. Equalization and proportioning. *Ind. Waste Treat.* 45–51. doi:10.1016/b978-
1092 012372493-9/50039-9

1093 Oubelkacem, A., Scardigno, A., Choukr-Allah, R., 2020. Treated Wastewater Reuse on Citrus in
1094 Morocco: Assessing the Economic Feasibility of Irrigation and Nutrient Management
1095 Strategies. *Integr. Environ. Assess. Manag.* 16, 898–909. doi:10.1002/ieam.4314

1096 Palazzolo, E., Laudicina, V.A., Rocuzzo, G., Allegra, M., Torrisi, B., Micalizzi, A., Badalucco, L.,
1097 2019. Bioindicators and nutrient availability through whole soil profile under orange groves
1098 after long-term different organic fertilizations. *SN Appl. Sci.* 1, 1–11. doi:10.1007/s42452-
1099 019-0479-3

1100 Parish, M.E., Braddock, R.J., Graumlich, T.R., 1986. Chemical and Microbial Characterization of
1101 Citrus Oil - Mill Effluent. *J. Food Sci.* 51, 431–433. doi:10.1111/j.1365-2621.1986.tb11148.x

1102 Qin, J., Enya, O., Lin, C., 2018. Dynamics of Fe, Mn, and Al liberated from contaminated soil by

1103 low-molecular-weight organic acids and their effects on the release of soil-borne trace
1104 elements. *Appl. Sci.* 8 (12), 2444. doi:10.3390/app8122444

1105 Regulation EU 2020/741, 2020. Regulation (EU) 2020/741, Minimum requirements for water reuse.
1106 Available online: [https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%](https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32019R1009)
1107 [3A32019R1009](https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32019R1009).

1108 Ren, C., Liu, K., Dou, P., Li, J., Wang, K., 2022. The Changes in Soil Microorganisms and Soil
1109 Chemical Properties Affect the Heterogeneity and Stability of Soil Aggregates before and after
1110 Grassland Conversion. *Agriculture* 12, 307. doi:10.3390/agriculture12020307

1111 Rosas-Mendoza, E.S., Méndez-Contreras, J.M., Aguilar-Lasserre, A.A., Vallejo-Cantú, N.A.,
1112 Alvarado-Lassman, A., 2020. Evaluation of bioenergy potential from citrus effluents through
1113 anaerobic digestion. *J. Clean. Prod.* 254, 1–11. doi:10.1016/j.jclepro.2020.120128

1114 Rosas-Mendoza, E.S., Méndez-Contreras, J.M., Martínez-Sibaja, A., Vallejo-Cantú, N.A.,
1115 Alvarado-Lassman, A., 2018. Anaerobic digestion of citrus industry effluents using an
1116 Anaerobic Hybrid Reactor. *Clean Technol. Environ. Policy* 20, 1387–1397.
1117 doi:10.1007/s10098-017-1483-1

1118 Saadatinavaz, F., Karimi, K., Denayer, J.F.M., 2021. Hydrothermal pretreatment: An efficient
1119 process for improvement of biobutanol, biohydrogen, and biogas production from orange
1120 waste via a biorefinery approach. *Bioresour. Technol.* 341, 125834.
1121 doi:10.1016/j.biortech.2021.125834

1122 Schimmenti, E., Borsellino, V., Galati, A., 2013. Growth of citrus production among the Euro-
1123 Mediterranean countries: Political implications and empirical findings. *Spanish J. Agric. Res.*
1124 11, 561–577. doi:10.5424/sjar/20131113-3422

1125 Sharma, K., Mahato, N., Cho, M.H., Lee, Y.R., 2017. Converting citrus wastes into value-added
1126 products: Economic and environmentally friendly approaches. *Nutrition* 34, 29–46.
1127 doi:10.1016/j.nut.2016.09.006

1128 Shen, S., Cheng, H., Liu, Y., Chen, Y., Chen, S., Liu, D., Ye, X., Chen, J., 2021. New electrolyte

1129 beverages prepared by the citrus canning processing water through chemical improvement.
1130 Food Chem. X 12, 100155. doi:10.1016/j.fochx.2021.100155

1131 Shrivastava, V., Ali, I., Marjub, M.M., Rene, E.R., Soto, A.M.F., 2022. Wastewater in the food
1132 industry: Treatment technologies and reuse potential. Chemosphere 293.
1133 doi:10.1016/j.chemosphere.2022.133553

1134 Strazzera, G., Battista, F., Garcia, N.H., Frison, N., Bolzonella, D., 2018. Volatile fatty acids
1135 production from food wastes for biorefinery platforms: A review. J. Environ. Manage. 226,
1136 278–288. doi:10.1016/j.jenvman.2018.08.039

1137 Suri, S., Singh, A., Nema, P.K., 2020. Current Applications of Citrus Fruit Processing Waste: A
1138 Scientific Outlook. Waste Manag. 11, 1–11. doi:10.1021/acssuschemeng.0c01735

1139 Tamburino, V., Zema, D.A., Zimbone, S.M., 2007. Depuration Processes of Citrus Wastewater, in:
1140 CIGR Section VI International Symposium on Food and Agricultural Products: Processing and
1141 Innovations.

1142 Torquato, L.D.M., Pachiega, R., Crespi, M.S., Nespeca, M.G., de Oliveira, J.E., Maintinguer, S.I.,
1143 2017. Potential of biohydrogen production from effluents of citrus processing industry using
1144 anaerobic bacteria from sewage sludge. Waste Manag. 59, 181–193.
1145 doi:10.1016/j.wasman.2016.10.047

1146 Ungureanu, N., Vlăduț, V., Voicu, G., 2020. Water scarcity and wastewater reuse in crop irrigation.
1147 Sustain. 12, 1–19. doi:10.3390/su12219055

1148 Vaish, B., Srivastava, V., Singh, P.K., Singh, P., Singh, R.P., 2020. Energy and nutrient recovery
1149 from agro-wastes: Rethinking their potential possibilities. Environ. Eng. Res. 25, 623–637.
1150 doi:10.4491/eer.2019.269

1151 Valentino, F., Morgan-Sagastume, F., Fraraccio, S., Corsi, G., Zanaroli, G., Werker, A., Majone,
1152 M., 2015. Sludge minimization in municipal wastewater treatment by polyhydroxyalkanoate
1153 (PHA) production. Environ. Sci. Pollut. Res. 22, 7281–7294. doi:10.1007/s11356-014-3268-y

1154 Violante, A., Cozzolino, V., Perelomov, L., Caporale, A.G., Pigna, M., 2010. Mobility and

1155 bioavailability of heavy metals and metalloids in soil environments. *J. Soil Sci. Plant Nutr.* 10,
1156 268–292. doi:10.4067/S0718-95162010000100005

1157 Wanner, J., 2017. *Activated sludge separation problems*, 2nd Edition. ISBN 9781780408637.
1158 doi:10.2166/9781780408644_053

1159 Yadav, V., Sarker, A., Yadav, A., Miftah, A.O., Bilal, M., Iqbal, H.M.N., 2022. Integrated
1160 biorefinery approach to valorize citrus waste: A sustainable solution for resource recovery and
1161 environmental management. *Chemosphere* 293, 133459.
1162 doi:10.1016/j.chemosphere.2021.133459

1163 Zema, D.A., Andiloro, S., Bombino, G., Caridi, A., Sidari, R., Tamburino, V., 2016. Comparing
1164 Different Schemes of Agricultural Wastewater Lagooning: Depuration Performance and
1165 Microbiological Characteristics. *Water. Air. Soil Pollut.* 227. doi:10.1007/s11270-016-3132-4

1166 Zema, D.A., Andiloro, S., Bombino, G., Tamburino, V., Sidari, R., Caridi, A., 2012. Depuration in
1167 aerated ponds of citrus processing wastewater with a high concentration of essential oils.
1168 *Environ. Technol. (United Kingdom)* 33, 1255–1260. doi:10.1080/09593330.2011.618938

1169 Zema, D.A., Calabro, P.S., Folino, A., Tamburino, V., Zappia, G., Zimbone, S.M., 2019.
1170 Wastewater management in citrus processing industries: An overview of advantages and
1171 limits. *Water (Switzerland)* 11, 2481. doi:10.3390/w11122481

1172 Zema, D.A., Calabrò, P.S., Folino, A., Tamburino, V., Zappia, G., Zimbone, S.M., 2018.
1173 Valorisation of citrus processing waste: A review. *Waste Manag.* 80, 252–273.
1174 doi:10.1016/j.wasman.2018.09.024

1175 Zhu, Ya, Zhai, Y., Li, S., Liu, Xiangmin, Wang, B., Liu, Xiaoping, Fan, Y., Shi, H., Li, C., Zhu,
1176 Yun, 2022. Thermal treatment of sewage sludge: A comparative review of the conversion
1177 principle, recovery methods and bioavailability-predicting of phosphorus. *Chemosphere* 291,
1178 133053. doi:10.1016/j.chemosphere.2021.133053

1179