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Intensified fish farming. Performance of electrochemical remediation of marine RAS waters

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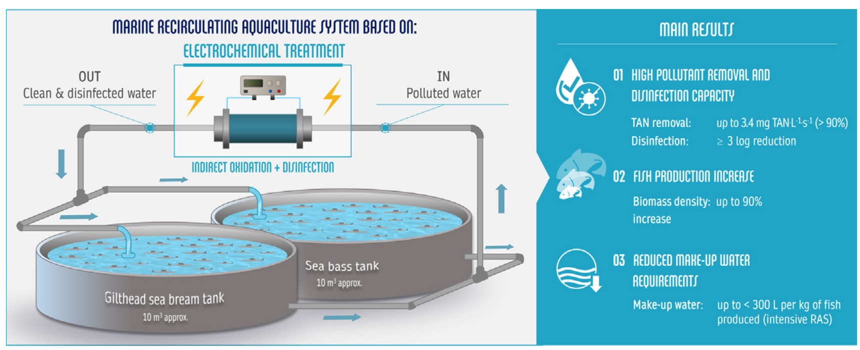
HIGHLIGHTS

- Current water treatment technology in RAS (biofiltration) has limited efficiency.
- Electrochemical oxidation has been tested as an alternative to biofiltration.
- High efficiency in TAN removal and simultaneous disinfection have been observed.
- Reduced water use (up to intensive RAS) and enhanced production have been achieved.
- Electrochemical oxidation can improve the environmental profile in aquaculture.

GRAPHICAL ABSTRACT



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ABSTRACT

Aquaculture has been the fastest growing agricultural sector in the past few decades and currently supplies about half of the fish market. A range of environmental and management concerns including limited land and water availability have led to intensifying fish production by recirculating aquaculture systems (RAS). Fish's diet contains 30–60 % protein and about 4–10 % nitrogen (N). As fish assimilate only 20–30 % of the feed to produce body mass, the unassimilated N is released in the form of toxic ammonium that deteriorates water quality and compels its degradation. Widely extended biological nitrification is not efficient in the removal of nitrites nor other chemicals and pharmaceuticals used during fish culture. Electrochemical oxidation, a less developed alternative, reports several advantages such as, i) simultaneous degradation of ammonia-nitrogen (TAN) and water disinfection in the same step with considerable simplification of the whole process, ii) easy adaptability to different production scales and periods of fish growth, and iii) no generation of harmful by-products and no use of chemicals, among others. Besides, in the case of marine aquaculture, the technology benefits from the high conductivity of seawater; thus, electrochemical oxidation is positioned in a very good place to satisfy the water treatment needs of the increasing production rate of marine aquaculture fish. Here, we report the analysis of the performance of a RAS demonstration plant aimed at farming gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*) and provided with electrochemical remediation of culture water. The performance of the plant, with 20 m^3 of seawater operating at a recirculation rate of 0.9–1.4 h^{-1}, has been analysed in terms of TAN removal, water disinfection, make-up water intake and energy consumption and compared to data of conventional RAS provided with biofilters. The benefits and advantages of the innovative electrochemical remediation of RAS water are highlighted.

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1. Introduction

The growing demand for fish, along with the dwindling natural fish populations and the necessity to reduce capture fisheries, has resulted in the increasing dependence on aquaculture as a source of fish for food. Indeed, aquaculture has been the fastest growing agricultural sector in the past few decades and currently supplies about half of the fish market (Mateo-Sagasta et al., 2017). A range of environmental and management concerns including limited land and water availability have led to intensifying fish production by recirculating aquaculture systems (RAS). As RAS holds a range of benefits over extensive aquaculture production (e.g., high fish yield, reuse of water, limited water exchange that reduces the release of contaminants to the environment, and less required land), it is expected to dramatically expand in the next few decades (Kouba et al., 2018; Timmons and Ebeling, 2013). Fish's diet contains 30–60 % protein and about 4–10 % nitrogen (N). As fish assimilate only 20–30 % of the feed to produce body mass, the unassimilated N is released in the form of toxic ammonium (NH_4^+) (Timmons and Ebeling, 2013). In an aqueous solution, ammonia (NH_3) and ammonium are in a pH-dependent equilibrium (Eq. 1),

$$NH_4{}^+ + OH^- \leftrightarrow NH_3 + H_2O \; pK_a = 9.24 \tag{1}$$

Currently, water treatment in RAS relies on biological nitrification to convert ammonia to nitrate, which is 100–200 folds less toxic. Nitrite is removed by dilution through daily water exchanges. Other contaminants (e.g., suspended solids) are also separated from the recirculating water and similarly removed. Such practice causes environmental contamination due to the discharge of N-containing wastewater, disposal of a valuable N which could be used for fertilization, and discharge of nitric oxide (N_2O), a known greenhouse gas which is produced in the nitrification process (Yogev et al., 2018; Hu et al., 2012). Furthermore, as nitrification relies on bacterial activity, ammonia removal rate is influenced by temperature and other chemicals/pharmaceuticals applied to the fish, e.g., antibiotics that are used against bacterial infections. This situation has promoted the research of different remediation alternatives for ammonium removal and recovery by non-biological methods, including direct and indirect oxidation (Gendel and Lahav, 2013; Díaz et al., 2011), adsorption on zeolites and activated carbon (Halim et al., 2010), and air stripping (Guštin and Marinšek-Logar, 2011). In general, these processes must face different drawbacks, such as high energy demand and/or application of chemicals that need further investigation to be solved. Other approaches such as reverse osmosis membranes struggle with low selectivity and high energy costs (Hurtado and Cancino-Madariaga, 2014).

The high salinity and conductivity of marine aquaculture waters leverages the application of electrochemical technologies, offering an ideal niche for this efficient water remediation technology. The performance of electro-oxidation processes relies strongly on the anode material, because it determines the type of electrochemical oxidants that are generated in the treated waters (Lacasa et al., 2012) and influences the effectiveness of active chlorine generation (Jeong et al., 2009). Successful results of the laboratory scale indirect electro-oxidation of ammonia have been previously reported in different aqueous media using boron doped diamond (Pérez et al., 2012; Anglada et al., 2010), and RuO_2-Ti, a more cost-effective material that reported high removal yield (Romano et al., 2020; Díaz, 2013).

In the electrochemical treatment of RAS water, total ammonia nitrogen (TAN) (NH_4^+/NH_3) oxidation occurs through the electrogeneration of chlorine (Cl_2), by direct oxidation of chloride on the anode, which is later on hydrolysed to form hypochlorous acid ($HOCl/OCl^-$), and then it reacts with TAN in a similar way to the breakpoint chlorination mechanism. The design of the remediation process relies on the kinetics of TAN oxidation, accounting for the phenomena that take place inside the electrochemical cell. Following the pioneering works focused on the kinetic study of TAN electro-oxidation as part of a remediation process of landfill leachates (Cabeza et al., 2007; Anglada et al., 2009), fundamental studies have been extended to ammonia oxidation in marine aquaculture waters (Ruan et al., 2016; Ding et al., 2015; Díaz et al., 2011; Li and Liu, 2009; Chen et al., 2007). Recently, Romano et al. (2021) collected previous information and reported a thorough analysis of the kinetics of ammonia oxidation in saline waters providing a useful tool for process design and optimization. Integrating the kinetics of direct anodic oxidation of chloride occurring at the electrochemical reactor, and ammonia chlorination reactions in the liquid bulk, facilitated the comprehension of the stages controlling the overall process kinetics and explained the pseudo zero-th order kinetic regime of TAN electrochemical removal observed in the low ammonia concentration range of interest in marine RAS systems.

Energy consumption together with the efficiency of water remediation are the main challenges facing the wide deployment of the technology. Romano et al. (2020) reported the comparison of the energy consumed considering different process configurations; in that work the authors highlighted that a trade-off between the minimum water uptake and the energy consumed would result in the minimum environmental impact.

Here, we present the design and performance of a demonstration pilot based on the electrochemical oxidation of ammonium-nitrogen for remediation of marine RAS waters in the farming of gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*). These are two marine species of special economic interest within the aquaculture sector, particularly in Europe, where they are included in the top 5 of most relevant species based on production (APROMAR, 2021).

The technical alternative here reported meets the standards to be used in marine RAS with protection of the environmental resources and minimum consumption of energy; next, substituting the energy source by higher percentage of renewable energy would increase the process sustainability and secure food for a larger population. Thus, this work constitutes a step forward in the deployment of more sustainable alternatives to secure fish for the growing population.

2. Materials and methods

2.1. Electrochemical oxidation of marine RAS water: fundamentals and process description

TAN is the key parameter to be addressed in the assessment of RAS as it is the main compound generated in the fish farming. In RAS, TAN can be rapidly accumulated, reaching unacceptable limits in a very short time if a sufficiently efficient treatment is not applied. In the conventional treatment of RAS water in biofilters, nitrites and nitrates are formed as a result of the nitrification process applied to remove TAN. Unlike a conventional RAS, the RAS based on electrochemical oxidation transforms TAN into nitrogen gas, minimizing the formation of nitrites and nitrates. Fig. 1 depicts the conversion of TAN in nitrogen through homogeneous reactions with electrogenerated chlorine in the liquid bulk.

When the electrochemical oxidation of TAN is followed the system requires additional steps as depicted in Fig. 2. Specifically, the RAS based on electrochemical oxidation studied in this work—under the name 'ELOXIRAS®' (APRIA Systems, S.L.)—integrated three stages, as follows:

i) Pre-treatment. Aimed at the removal of solid particles, oils and fats. First, the untreated water flows by gravity from the farming tanks to a drum filter where coarse solid particles are removed. Then, water flows to a buffer tank that acts as a water reservoir, and oils and fats interceptor. Fine solids and organic nitrogen are removed by passing a fraction of the water through a foam fractionator. Finally, the clarified water is pumped to a filtration sub-stage using activated glass filtration media.
ii) Main treatment. The aim is the removal of pollutants and pathogens by means of electrochemical oxidation. Water is pumped from the pre-filters to the electrochemical unit, which consists of four electrochemical cells—each powered by their respective power source—arranged in parallel, so they can be used simultaneously or alternately, as required. Each cell consists of a 6-inch PVC-U tubular casing that contains 5 electrode packages; the total effective anode surface area of each cell is

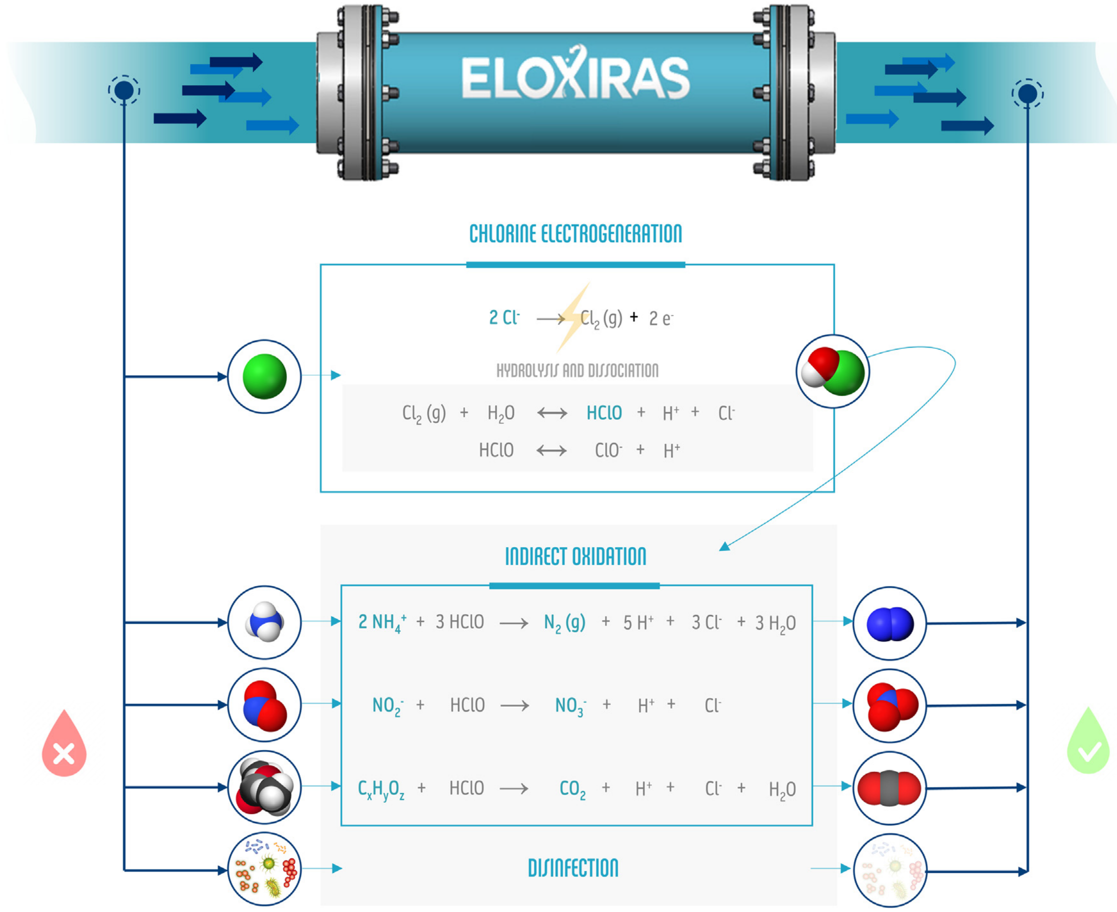

Fig. 1. Main reactions involved in the electrochemical treatment of marine aquaculture water in RAS.

$0.57\ m^2$. Each electrode package consists of 5 pairs of rectangular bipolar electrodes made of titanium coated with ruthenium oxide as catalyst (RuO_2-Ti). The current intensity supplied by the power sources is divided between the different electrode packages. As a result, a mixture of oxidants as chlorine derivatives are electrogenerated from the chlorides present in the water—a minimum salinity of 10 $g{\cdot}L^{-1}$ is required; the influence of salinity in the electro-generation of chlorine is presented in Fig. S1 in Supplementary Material—and indirect oxidation reactions take place, removing TAN.

iii) Post-treatment. Aimed at the removal of undesired by-products of the electrochemical process, to ensure an optimal water quality. These by-products are: (i) residual total chlorine—i.e., the sum of residual free chlorine (excess chlorine that has not been consumed in the treatment) and combined chlorine (product of the bond between chlorine not consumed in the treatment and other substances, usually of organic nature), (ii) disinfection by-products (DBPs) in the form of trihalomethanes (THMs) resulting from the chlorination, and (iii) electro-generated halogenated ions—chlorates, perchlorates and bromates—due to electrochemical side reactions with halides present in seawater—i.e., chlorides and bromides. The post-treatment consists of two sub-stages: (i) adsorption by means of granular activated carbon (AC), and (ii) afterwards, gas balance by means of a degassing unit where N_2, CO_2, CO, H_2, and other potential gases are stripped out by flowing air counter-currently. Then, the water returns to the farming tanks by gravity.

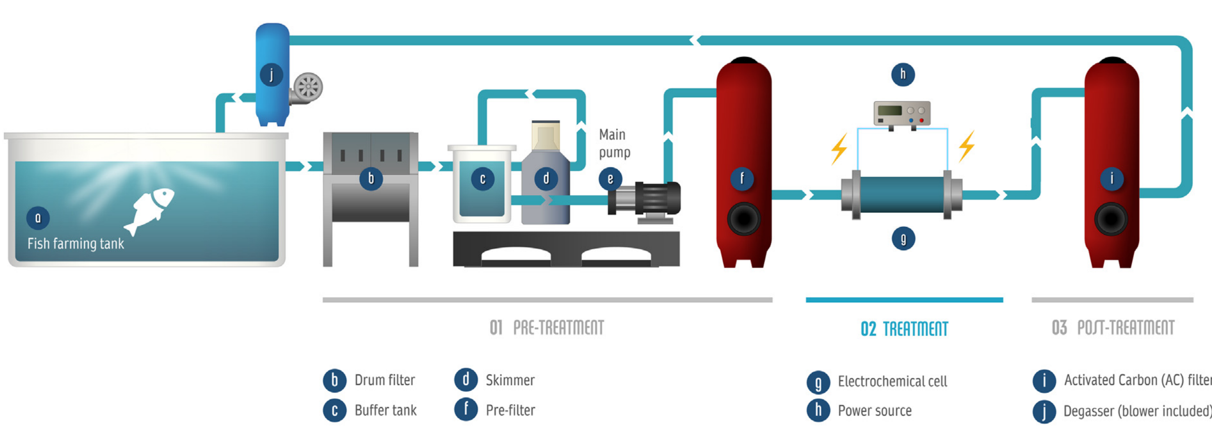

Fig. 2. Schematic representation of the RAS based on electrochemical treatment tested in the present study.

2.2. Assessment methods

The system incorporated automatic on-line analysers for monitoring chemical and physical parameters relevant to aquaculture—i.e., pH, dissolved oxygen and temperature, as well as specific variables of the electrochemical treatment, namely, oxidation-reduction potential, TAN and total chlorine. The TAN analyser (Instrumentación Analítica, S.A.), by means of a gas-sensing ion selective electrode, provided TAN concentration in real time. Periodic monitoring of other relevant compounds was carried out through the analytical measurement of nitrites, nitrates, and chemical oxygen demand (COD). Nitrogen derivatives were determined by photometry, using a multiparametric photometer HI83303 (Hanna Instruments®, S.L.). COD was determined by titration following the heat of dilution method proposed by Ruttanagosrigit and Boyd (1989) for waters with high chloride concentration.

In addition, the disinfection yield in the treated water was also checked. In this regard, it is important to focus on those bacteria that are a source of frequent diseases affecting the farming of the studied fish species. In this sense, *Vibrio* spp. are the most common and serious pathogens in fish and shellfish marine aquaculture worldwide, which cause a disease known as vibriosis (Muniesa et al., 2020; Mohamad et al., 2019)—specifically, the subspecies *V. anguillarum* (FAO, 2022a; Frans et al., 2011; Korum and Timur, 2008; Breuil and Haffner, 1989) and *V. damsela* (FAO, 2022b; Abdel-Aziz et al., 2013) are responsible for vibriosis in sea bass and gilthead sea bream farming, respectively. Therefore, analyses of total bacteria and *Vibrio* spp. were performed. The determination of the colony forming units (CFU) was conducted by means of an adaptation of the standard plate count method APHA 9260 using tryptone soy agar (TSA) and tryptone citrate bile sucrose agar (TCBS) as growth media for total colonies and total Vibrionaceae counts, respectively. As required, the analytical determinations were performed immediately after sampling to avoid contamination. Chlorinated samples at the outlet of the electrochemical unit were neutralized with sodium thiosulfate.

The assessment of water quality run in parallel to the control of fish production. To this end, typical growth and feeding indicators were considered, namely:

Specific Growth Rate (*SGR*). It relies on the absolute growth. Results are given in percentage increase per day, according to Eq. 2,

$$SGR = \frac{\log\left(w_t/w_i\right)}{t} \times 100 \tag{2}$$

where w_t and w_i are the final and initial average fish weight, respectively, and t is the farming period in days.

Feed Conversion Ratio (*FCR*). It measures the efficiency of the conversion of feed into new fish biomass: the amount of feed supplied compared to the amount of biomass generated. Accordingly:

$$FCR = \frac{\textit{Total feed supplied}\ (\text{kg})}{\textit{Final biomass}\ (\text{kg}) - \textit{Initial biomass}\ (\text{kg})} \tag{3}$$

From Eq. 3, it is clear that lower *FCR* values indicate efficient conversion and, thus, they are desirable.

Specific Feed Rate (*SFR*). The *SFR* is calculated as the amount of feed that is supplied daily per amount of biomass (Eq. 4).

$$SFR = \left[\left(\frac{\textit{Total feed supplied}(\text{kg})}{t(\text{days})}\right) / \left(\frac{\textit{Initial biomass}(\text{kg}) + \textit{Final biomass}(\text{kg})}{2}\right)\right] \times 100 \tag{4}$$

The *SFR* is a percentage fraction of the fish weight and is closely related to the *SGR* and *FCR* indexes because every fish species has its own standard growth potential at a certain size and, if more feed than required is supplied then it would be wasted, increasing the *FCR* value accordingly. Section 3.2.1 of the present manuscript includes typical values of these indicators for the species studied in this work depending on their growth stage, as reported in the literature (Ortega, 2013; Ortega, 2008).

3. Case study

3.1. Farming of gilthead sea bream (Sparus aurata) and sea bass (Dicentrarchus labrax)

The present case of study reports the performance of a marine RAS for the simultaneous farming of two fish species—gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*)—provided with electrochemical oxidation technology—ELOXIRAS®—of water. The performance of a demonstration plant under real operating conditions was conducted in an aquaculture facility located in Tarragona (Spain). A picture of the demonstration plant can be found in Fig. S2 in the Supplementary Material. This case of study, which involves the farming under a real scenario of 2 species of great economic interest, was selected as it was considered to be representative enough to demonstrate the general applicability of the electrochemical oxidation technology in RAS for marine aquaculture.

Fish farming of gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*) included different farming stages—pre-growing and on-growing—and different stocking conditions—standard stocking and high production stocking. The different essays performed were conducted using two separate fish farming tanks of approximately 12 m^3 of maximum water capacity each—one tank for each species. Both tanks were connected hydraulically to the same RAS based on electrochemical oxidation; thus, water from the two tanks—with an average salinity level of 37 $g \cdot L^{-1}$—was treated undivided. The standard pre-growing essay started with 9728 individuals of gilthead sea bream and 13,506 individuals of sea bass with an average specimen weight of 15.46 ± 3.47 g and 12.18 ± 3.41 g, respectively. The same specimens of fish were used for the standard on-growing essay. Similarly, the high production pre-growing essay started with 26,473 new individuals of gilthead sea bream and 24,213 new individuals of sea bass, with an average weight of 6.27 ± 1.70 g and 9.11 ± 2.14 g, respectively. The same specimens of fish were used for the high production on-growing essay. During and between the different essays, the amount of fish biomass was biometrically measured, and fish specimens were removed periodically from the fish tanks in order to maintain the biomass density within the range required for the tests. All the tests were carried out in accordance with the guidelines provided for animal protection by the European Union (Directive 2010/63/EU, 2010).

Table 1 reports a brief description of the main farming conditions in relation to the different demonstration tests conducted. Total rearing water volume and fish biomass were selected to test the RAS based on electrochemical oxidation under large scale operation conditions, and, also, for testing the operation at high biomass—stocking—density. All demonstration tests were conducted under variable operating conditions, adjusting the electrochemical treatment intensity as required. Average operating conditions, including global treatment flowrate, recirculation rate—number of times that the total culture volume flows through the system per unit of time, and current density applied for the electrochemical treatment, are presented as well in Table 1.

3.2. Performance of the electrochemical remediation of RAS waters

Next, the results related to the performance of the electrochemical remediation of RAS water in terms of TAN reduction and water disinfection are presented and discussed.

3.2.1. Control of total ammonia nitrogen

An intensive analytical monitoring was carried out to check and optimize the removal of nitrogen compounds, with focus on the efficacy of TAN removal. In this regard, it must be considered that the generation rate of TAN is dependent on the farming conditions—i.e., biomass density, fish size and feed. In addition, for a given level of TAN, the removal rate is determined by the operating conditions of the treatment—i.e., flowrate and current density. In this sense, the electrochemical oxidation technology allows to easily modulate the removal rate of TAN by adjusting the treatment intensity through these operating variables. High TAN removal rates can be

Table 1
Farming conditions in the demonstration tests of ELOXIRAS® (rearing water volume: 20 m^3 approx.).

Species	Farming conditions								Average operating conditions	
	Farming stage	Stocking conditions	Duration (weeks)	Culture vol. (m^3)	Biomass density ($kg·m^{-3}$)			Initial size (g)	Total flowrate[b] ($m^3·h^{-1}$)	Current density ($A·m^{-2}$)
					Typical[a]	Initial	Avg.			
Gilthead sea bream	Pre-growing	Standard	10	9.40	<20	16.0	20.1	15.46	30.5	217.6
		High production	5	10.92	N/A	15.2	29.4	6.27	24.8	418.0
	On-growing	Standard	25	10.92	<35	22.1	24.7	57.64	23.5	271.9
		High production	5		N/A	31.6	41.2	24.36	20.2	451.8
Sea bass	Pre-growing	Standard	10	9.40	<20	17.5	22.5	12.18	30.5	217.6
		High production	5	10.92	N/A	20.2	30.0	9.11	24.8	418.0
	On-growing	Standard	25	10.92	<23	20.7	23.4	34.70	23.5	271.9
		High production	5		N/A	30.6	38.3	31.36	20.2	451.8

[a] N/A: not applicable.
[b] Corresponding average recirculation rate: 1.4 h^{-1} (standard pre-growing), 1.1 h^{-1} (high production pre-growing), 1.1 h^{-1} (standard on-growing), and 0.9 h^{-1} (high production on-growing).

achieved through this adjustment. In particular, based on the best results obtained in the present study, the electrochemical oxidation technology is able to remove TAN up to a rate of 3.4 $mg·L^{-1}·s^{-1}$ (the unit of volume is referred to the volume of the electrochemical cells and the unit of time is referred to the residence time inside the electrochemical cells), with an efficacy higher than 90 %—operating conditions: total flow rate, Q_{total} = 24.8 $m^3·h^{-1}$ (using 2 electrochemical cells; 12.4 $m^3·h^{-1}$ per cell), current density, j = 701.8 $A·m^{-2}$; initial concentrations: $[TAN]_0$ = 1.83 mg $TAN·L^{-1}$, $[NO_2^-]_0 = 1.58 \cdot 10^{-1}$ mg $N\text{-}NO_2^-·L^{-1}$, $[COD]_0$ = 1.8 mg $O_2·L^{-1}$. Typical values reported in the literature for the TAN removal rate associated with biofiltration technology (conventional treatment in RAS) are referred to specific variables involved in the technology (e.g., specific surface area or volume of the filtration media) and, therefore, are not directly comparable. In addition, TAN removal efficacy is dependent on many factors, such as the scale of the system (related to the total production and the total culture volume), the farming conditions (i.e., species, rearing stage, biomass or stocking density, etc.), and/or the treatment conditions (i.e., treatment flow rate, number of daily recirculation of the culture volume, etc.). Based on that, for a proper comparison, TAN removal efficacy must be assessed using an indicator that may be expressed in terms that are: (i) common to both technologies, and (ii) as independent as possible from the farming and treatment conditions. For that reason, TAN removal efficacy should be compared through indicators such as the TAN removal rate (expressed in equivalent units: e.g., as $mg·L^{-1}·s^{-1}$), TAN removal capacity (e.g., $mg·m^{-3}$ of treated water), or percentage TAN removal.

A review on the TAN removal efficiency of the main types of treatment systems among those based on biofiltration technology aimed at their application in the aquaculture sector has been performed. Only studies conducted under conditions equivalent to those applied in this study—i.e., <2 passes of the total rearing volume through the treatment per hour, stocking densities lower than 50 $kg·m^{-3}$, and inlet concentrations of TAN lower than 4.0 $mg·L^{-1}$)—have been considered (Stanwat et al., 2020; Keuter et al., 2017; Godoy-Olmos et al., 2016; Wahyuningsih et al., 2015; Liu et al., 2013; Díaz et al., 2012; Harwanto et al., 2011; Kumar et al., 2011; Kumar et al., 2010; Suhr and Pedersen, 2010; Brazil, 2006; Tseng and Wu, 2004; Sandu et al., 2002; Miller and Libey, 1985). The most relevant information is summarized in Table S1 in Supplementary Material. Based on that review, from data calculated by the authors from the reported technical information, the most frequent types of biofilters are expected to remove TAN at a rate up to 0.028 $mg·L^{-1}·s^{-1}$. This value is far below 3.4 $mg·L^{-1}·s^{-1}$—the value obtained for the RAS based on electrochemical oxidation. That is due to the fact that biofiltration requires longer residence times than the electrochemical oxidation to reach an adequate TAN removal. As for the TAN removal capacity, it ranges 137.4–1426.5 mg $TAN·m^{-3}$ of treated water, with percent TAN removal indices lower than 90 %. Making the same calculation with the results obtained in the present study, the electrochemical alternative has proven to be able to reach a TAN removal efficacy up to 1670.0 mg $TAN·m^{-3}$ of treated water and a percent TAN removal higher than 90 % (both greater than typical values). Based on that, it has been demonstrated that electrochemical oxidation technology has a better performance for the management of TAN in marine RAS.

Nevertheless, the level of TAN fluctuates throughout the day due to feeding and the metabolic activity of the animals. For that reason, as the level of TAN decreases, lower removal rates are required to keep TAN under suitable limits. Based on the above, and on the experience acquired during the experimentation, in order to keep the level of TAN under control it is not always imperative to operate at conditions so intense as to achieve removal efficacies as high as the maximum observed. In this regard, if the influence of operating conditions on the efficacy of the electrochemical TAN removal process is known, the adaptability of the RAS based on electrochemical oxidation offers the possibility to optimize resources and consumptions as much as possible—modulating the intensity as required. In this sense, a study on the influence of current density on the removal rate of TAN was conducted. The results are summarized in Fig. S3 in the Supplementary Material.

Fig. 3 reports the time evolution of TAN concentration in the farming tanks for the different demonstration tests performed. As seen, it describes a characteristic saw-shaped line that reflects feeding periods, denoted by the peaks that appear as a consequence of the increased metabolic activity right after the feed intake. For a better visualization and distinction of the peaks, Fig. 3 is plotted for short selected periods within the full execution periods of the tests conducted.

From results in Fig. 3, it is clear that, for given stocking conditions, the generation rate of TAN is not particularly dependent on the size of the fish —TAN fluctuations under standard stocking conditions obtained for pre-growing and on-growing tests (Fig. 3a and b, respectively) are comparable to each other; the same is observed when high production stocking conditions are applied. In turn, for given species at a particular farming stage, the level of TAN—both basal and peak value—is intimately related to the biomass density. In particular, for the tests conducted under standard stocking conditions (Fig. 3a and b), basal values were around 0.25 mg $TAN·L^{-1}$ and regular peak values ranged from 0.5 to 1.5–2.0 mg $TAN·L^{-1}$; whereas for the homologous tests conducted under high production stocking conditions (Fig. 3c and d), basal values were around 0.5 mg $TAN·L^{-1}$ and regular peak values ranged from 2.0 to 3.5–4.0 mg $TAN·L^{-1}$. However, results evidence that, regardless of how high those peak values were, the RAS based on electrochemical oxidation proved to be able to pull down TAN

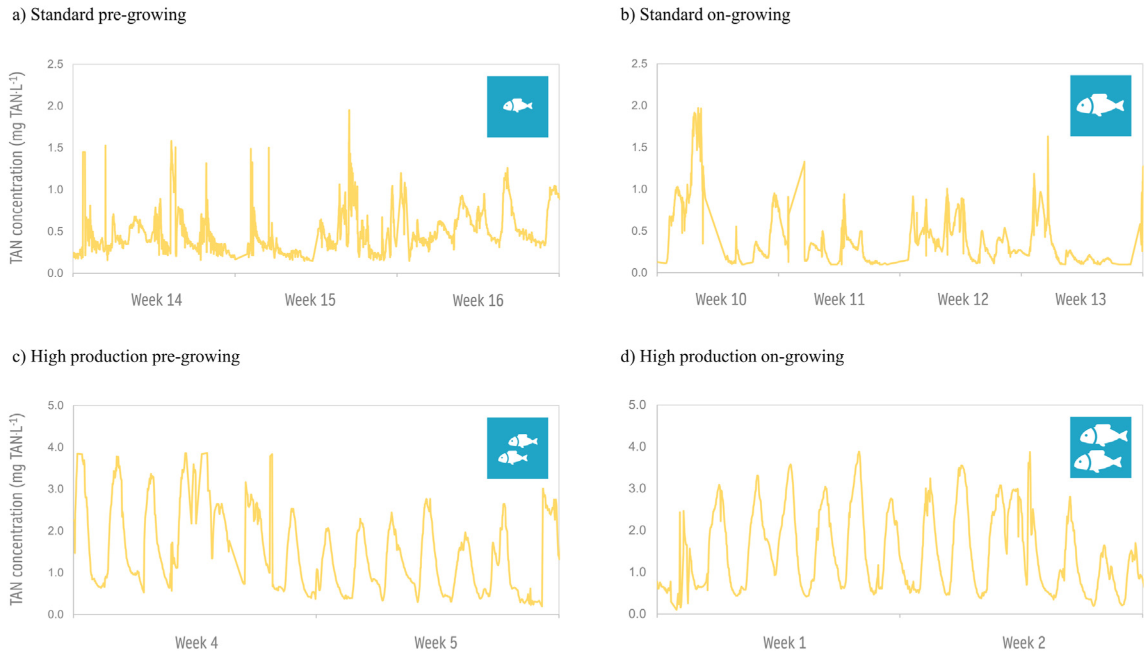

Fig. 3. Time evolution of TAN over selected periods within the different ELOXIRAS® demonstration tests conducted in RAS.

peaks, turning TAN back to the corresponding basal levels in a short period of time with no detrimental effect on the fish—only long-term exposures to values >3 mg TAN·L^{-1} may result in damages to warmwater fish health (Timmons and Ebeling, 2013).

3.2.2. Disinfection

Disinfection has been assessed by comparing the microbiological counts of total bacteria and *Vibrio* spp. both in the rearing water and in the outlet stream of the electrochemical treatment. In this sense, for the different demonstration tests performed, total bacteria and *Vibrio* spp. were found in the rearing water. Graphic evidence of the disinfection associated with the RAS based on electrochemical oxidation is provided in fig. S4 in the Supplementary Material, where the total bacteria and *Vibrio* spp. counts are presented for illustrative purposes; as seen, no significant growth is observed at the outlet of the electrochemical cell. In particular, removal efficacies >99.9 % were found for both total bacteria and *Vibrio* spp. in all cases. Furthermore, given that the levels of total bacteria in the rearing water were maintained in the order of 10^2–10^3 CFU·mL^{-1} over time, it is inferred that a disinfection capacity greater or equal to 3 log CFU·mL^{-1} is likely to be achieved in a RAS based on electrochemical oxidation. Overall, these results confirm the disinfection potential of the electrochemical oxidation technology. Moreover, they reveal that the disinfection capacity of ELOXIRAS®, UV or ozone treatment are equivalent (Summerfelt et al., 2009; Sharrer and Summerfelt, 2007).

Based on the above, it has been proved that RAS based on electrochemical oxidation is able to contribute to control the proliferation of concerning pathogens that are commonly found in marine recirculation aquaculture. In this sense, the quality of the rearing water in marine aquaculture facilities is guaranteed not only in terms of level of pollutants derived from the metabolism of the fish—i.e., TAN, nitrites or dissolved organic matter, but also in terms of microbial load. Hence, ELOXIRAS®, contributes to the stabilization of the microbiota of the culture tanks, a key issue required for the physical welfare of the farmed fish.

3.3. Assessment of the performance of fish production

Next, the assessment of the performance of fish production in terms of fish growth—related to the farming process economics—and physical welfare will be presented and discussed.

3.3.1. Fish growth

In commercial aquaculture facilities, the feeding and growth performance of fish specimens are the most influential factors regarding economic benefits and are quantified by means of widely employed indicators: specific growth rate—*SGR*, specific feed rate—*SFR*, and feed conversion ratio—*FCR*. The results obtained for each of these indicators for gilthead sea bream and sea bass are reported in Table 2, respectively. Data of maximum biomass density are also included in this table for contextualization purposes.

According to the results shown in Table 2, farming under standard stocking conditions at both pre-growing and on-growing stages is considered satisfactory. On the one hand, *SGR* values were not too far from estimated typical values—even slightly above for both gilthead sea bream and sea bass on-growing farming were obtained, which indicate an acceptable development of the specimens. On the other hand, *FCR* values were within typical ranges or slightly below, evidencing adequate use of feed. This demonstrates the capacity of the RAS based on electrochemical oxidation for farming under standard stocking conditions. In addition, Fig. S5 in Supplementary Material illustrates the growth observed for gilthead sea bream and sea bass as a result of the pre-growing demonstration tests conducted under standard stocking conditions.

With respect to high production stocking conditions, typical biomass densities of 10–20 kg·m^{-3} (Ortega, 2008; Ortega, 2013) for gilthead sea bream and sea bass pre-growing farming have been widely exceeded reaching 36.1 kg·m^{-3} (1.8-fold) and 38.6 kg·m^{-3} (1.9-fold), respectively, with no detrimental effect observed on the regular development of the farmed fish—specific growth rates of 5.1 %·day^{-1} (gilthead sea bream) and 4.3 %·day^{-1} (sea bass) have been achieved, nor on their appetite and use of feed—*SFR* values of 5.0 and 5.2 %·day^{-1}, within the typical range of 3.0–8.0 %·day^{-1}, were obtained for gilthead sea bream and sea bass, respectively; *FCR* values of 1.3 and 1.5 kg feed kg fish produced^{-1}, at the limits of the typical range of 1.1/1.2–1.4, were obtained for gilthead sea bream and sea bass, respectively. Furthermore, according to Ortega (2008, 2013), the maximum insurable biomass density values for the on-growing farming stage of gilthead sea bream and sea bass are 35 and 23 kg·m^{-3}, respectively. Maximum biomass density values of 48.9 kg·m^{-3} (1.4-fold) and 43.7 kg·m^{-3} (1.9-fold) were reached for gilthead sea bream and sea bass on-growing farming, respectively. In addition, the amount of feed consumed with respect to the fish size per day was

Table 2

Feeding and growth performance of gilthead sea bream and sea bass farming using RAS based electrochemical oxidation (ELOXIRAS®).

Parameter	Target	Pre-growing			On-growing		
		Typical values[a]	Standard production	High production	Typical values[a]	Standard production	High production
Gilthead sea bream							
Max. stocking density ($kg \cdot m^{-3}$)	–	20	24.8	36.1	35	25.2	48.9
SGR (%$\cdot day^{-1}$)	≥Typical values	3.8[b]	2.1	5.1	0.56[b]	1.1	2.0
FCR (kg feed/kg fish produced)	≤Typical values	1.2–1.4	1.0	1.3	1.5–2.0	1.3	2.1
SFR (%$\cdot day^{-1}$)	Within typical range	3.0–8.0	1.5	5.0	0.5–3.5	0.5	3.8
Sea bass							
Max. stocking density ($kg \cdot m^{-3}$)	–	20	28.6	38.6	23	28.9	43.7
SGR (%$\cdot day^{-1}$)	≥Typical values	3.8[c]	1.6	4.3	0.60[b]	1.2	1.6
FCR (kg feed/kg fish produced)	≤Typical values	1.1–1.4	1.2	1.5	1.5–2.0	1.5	2.6
SFR (%$\cdot day^{-1}$)	Within typical range	3.0–8.0	1.4	5.2	0.5–3.0	0.6	3.7

[a] Typical values according to Ortega (2008) and Ortega (2013) for gilthead sea bream and sea bass, respectively.
[b] Estimation calculated from data according to Ortega (2008).
[c] Estimation calculated from data according to Ortega (2013).

slightly greater than the maximum typical values—3.8 and 3.7 %$\cdot day^{-1}$ in comparison with a typical range of 0.5–3.0/3.5 %$\cdot day^{-1}$, that leads consequently to considerably increased growth rates—2.02 and 1.63 %$\cdot day^{-1}$ in contrast with typical values of 0.6 %$\cdot day^{-1}$. However, these increased growth rates were obtained at the expense of a negligible decay in the use of feed—2.1 and 2.6 kg of feed per kg of fish produced compared with typical values of 1.5–2—as is expected when fish undergo fast growth, given that the feed assimilation rate or growth potential of the organism of the animals cannot be surpassed. In spite of that, the overall performance under high production stocking conditions is still satisfactory as the increase in production—e.g., around 90 % increased production for sea bass on-growing with respect to typical values—offsets by far the extra feed consumed—e.g., from 30 % to 60 % (worst-case scenario) for sea bass on-growing with respect to typical values. This fact, together with a growth above average, evidence that the RAS based on electrochemical oxidation can duplicate the aquaculture productivity—by raising the biomass density almost twice as much—while maintaining the farming performance in terms of feeding and growth within a satisfactory level, according to typical reference values of usual performance indicators.

Additionally, in order to confirm the regular development and growth of fish, the evolution of the average size of the specimens was monitored throughout the entire duration of the tests and compared with that of a control group of fish consisting of specimens from the same batch. Benchmarking was conducted with a control group that was farmed in parallel in the same aquaculture facility following conventional RAS practices—that is, under standard stocking conditions and using a commercial RAS based on biofiltration instead of electrochemical oxidation. Fig. 4 shows the evolution of fish size observed for different tests performed compared to that of the control group.

As shown in Fig. 4a, under standard stocking conditions, the growth underwent by the specimens of both species when farmed using ELOXIRAS® was fairly similar to that observed in the specimens of the control group, farmed using a RAS based on biofiltration. This result supports the representativeness of the tests performed in this study for the demonstration of the RAS based on electrochemical water treatment alternative. The slight differences observed in the average size of fish could be attributed to the fact that, due to the limited space available, the farming demonstration tests using ELOXIRAS® were conducted outdoors, while those performed using the biofiltration-RAS were conducted indoors. Indoor farming allows better control of temperature, thus providing more constant values; on the other hand, outdoor farming entails a higher level of exposure to the environment and, therefore, it is more dependent on weather conditions and temperature variations between day and night cycles. In this sense, tests carried out during spring- and summertime provided more stable results,

Fig. 4. Gilthead sea bream and sea bass growth: (a) standard pre-growing test; (b) high production pre-growing test.

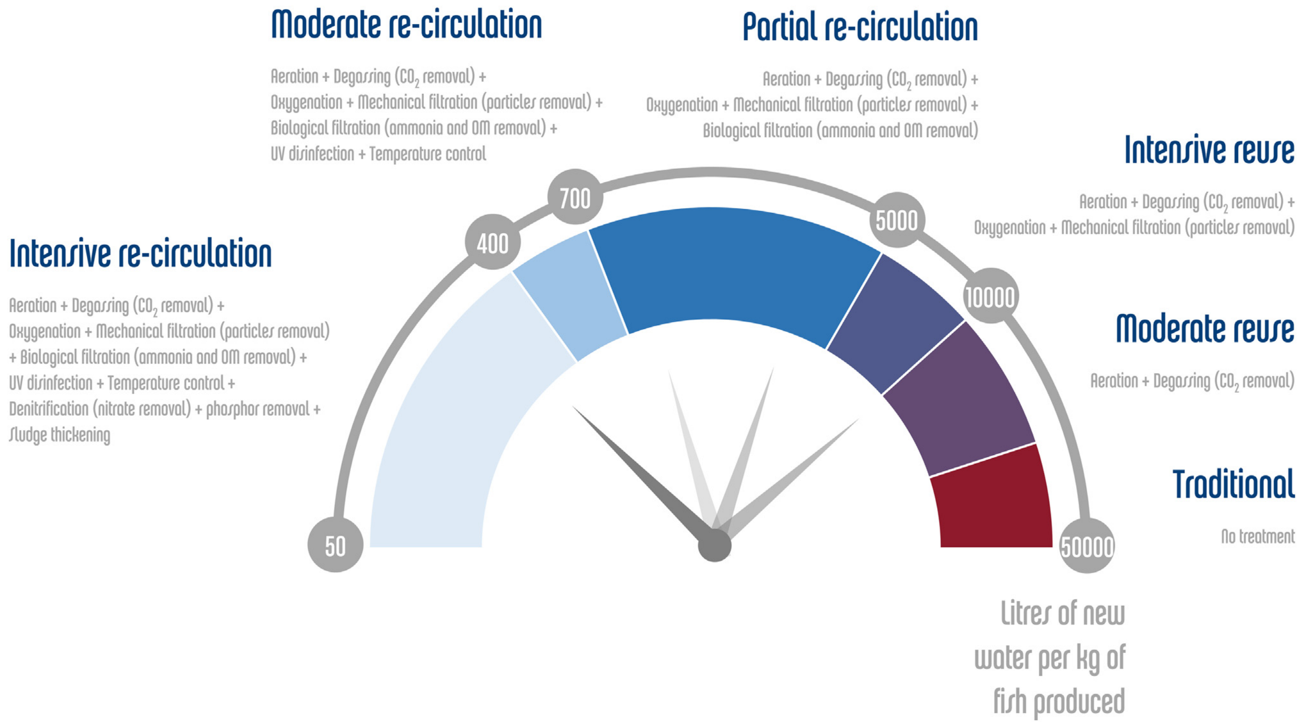

Fig. 5. Classification of fish farms based on water consumption in terms of make-up water intake (adapted from Heldbo, 2014).

when weather conditions helped to maintain constant temperature, even when they were conducted at high biomass densities—around 40 kg·m^{-3}. In fact, Fig. 4b shows an analogous or even a better growth in the tests performed in spring under high production conditions in spite of the increase in biomass with respect to that of the control group.

3.3.2. Fish health

The assessment of production from the point of view of physical welfare or health of fish was conducted by means of histopathological studies carried out by the Fish Diagnostic Service of Universitat Autònoma de Barcelona (Spain). In this study, the sampling of 16 specimens of gilthead sea bream and 16 specimens of sea bass was conducted according to common protocols for histopathological diagnosis in fish. The specimens were measured and weighted, and samples of gill, liver, spleen, kidney, intestine, heart, encephalon, digestive tract, skin, and musculature were fixed in formaldehyde for subsequent analysis.

The histopathological studies delivered very positive results. In the first place, regarding the overall examination of the fish specimens, they all were in very good condition and no macroscopic injuries were observed; except for some injuries in the fins, which are attributed to the capture process. In the second place, with respect to the examined organs, the quality of the samples was particularly good, allowing a detailed examination. As a result, no anomalies or injuries with pathological significance were found. It is noteworthy the absence of alterations typically observed in farmed fish—using either farming cages, flow-through systems, or RAS, such as small changes in gill, skin, or kidney tissues. These three organs are good indicators of the effects of water quality. Accordingly, it is advanced that ELOXIRAS® provides higher water quality than other farming alternatives.

3.4. Environmental performance

Next, the evaluation of the environmental performance in terms of resource consumption will be discussed. In this regard, it is worth noting that, when comparing the RAS based on electrochemical oxidation with other conventional RAS in terms of water and energy consumption, a conventional RAS that includes all possible extra treatment stages (i.e., disinfection and denitrification) must be selected as benchmark for comparison.

3.4.1. Water consumption

Regarding water consumption, a common classification of fish farms has been proposed, as shown in Fig. 5. Table 3 contains the data of water consumption observed for the electrochemical treatment of RAS waters, after the different demonstration tests performed. Based on the results obtained and considering the classification presented in Fig. 5, electrochemical oxidation achieved the category of intensive RAS under standard stocking conditions, with a make-up water intake lower than 300 L kg fish produced^{-1}. In addition, this result also evidences that the make-up

Table 3
Evaluation of water consumption of RAS based on electrochemical oxidation at large scale for gilthead sea bream and sea bass farming.

Farming stage	Stocking conditions	Avg. recirculation rate (h^{-1})	Gilthead sea bream/sea bass biomass density (kg·m^{-3})		Make-up water intake (L kg fish produced^{-1})
			Typical[a]	Max.	
Pre-growing	Standard	1.4	<20/<20	24.8/28.6	1100
	High production	1.1	N/A/N/A	36.1/38.6	1800
On-growing	Standard	1.1	<35/<23	25.2/28.9	270
	High production	0.9	N/A/N/A	48.9/43.7	700

[a] N/A: not applicable.

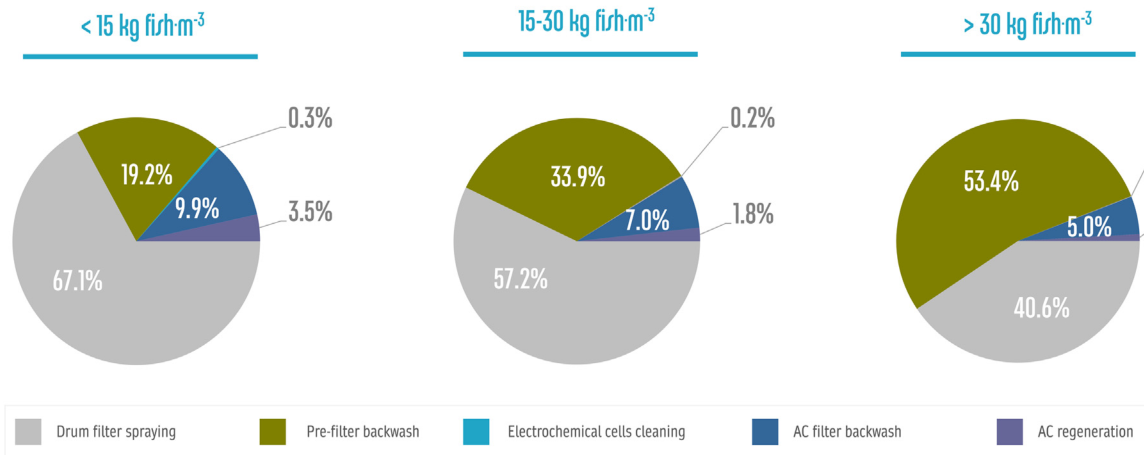

Fig. 6. Contribution of water-consuming operations of the RAS based on electrochemical oxidation to the total make-up water intake required at different biomass densities.

water requirements of ELOXIRAS® may be around 50 % lower than conventional RAS with comparable functionality—that is, disinfection included (at least); considering moderate RAS, 400–700 L new water kg fish produced^{-1} are usually required (Fig. 5).

As for high production stocking conditions—biomass densities up to 90 % higher than standard (even above 40 kg·m^{-3}), the minimum water consumption achieved was 700 L kg fish produced^{-1}. This value suggests that it is difficult to reach the intensive RAS category when high biomass densities are involved. Nonetheless, it is worth mentioning that, given high production stocking conditions, the water consumption is mainly due to the filtration stage included as part of the pre-treatment to control the level of suspended solids, and oils and fats, and, thus, unrelated to the electrochemical treatment itself—that means that the same would happen with any other alternative treatment available in the market if they were to be applied to farm fish at so high biomass densities. In this regard, Fig. 6 shows the distribution of make-up water consumption of RAS based on electrochemical oxidation by treatment stages and depending on the biomass density applied throughout the different demonstration tests. As shown, make-up water was consumed almost entirely in the cleaning operations required for the pre-treatment and post-treatment filters—drum filter spraying and pre-filters backwash (pre-treatment), and AC filter backwash and carbon regeneration (post-treatment), while no additional water was consumed specifically for the dilution of pollutants in the RAS farming circuit, unlike many conventional RAS. In this sense, dilution practices were not required because ELOXIRAS® is able to remove pollutants with very high efficacies and with no accumulation of undesirable by-products—the levels of the main by-products (total chlorine, THMs, and halogenated ions) were periodically monitored; the most relevant results are presented in Table S2 in Supplementary Material.

Pre-treatment itself was responsible for 86.3–94.0 % of the total water consumption (Fig. 6a–c), depending on the biomass density tested—the higher the biomass density, the higher the quantity of solids, oils, and fats generated, and, therefore, the higher the amount of water required by the pre-treatment. Thus, the main treatment and the post-treatment together account for just 6.0–13.7 % of the total make-up water uptake (Figs. 6a–c). All in all, the RAS based on electrochemical oxidation is still in the range of moderate RAS for high production stocking conditions.

3.4.2. Energy consumption

In order to assess the energy efficiency of the process—and to be able to properly compare it with conventional processes in terms of energy use, the energy consumption must be related to relevant cost and technical production variables; typically: the amount of feed supplied, the amount of TAN removed, or the volume of treated water.

Table 4 reports the energy consumption of ELOXIRAS® for the different demonstration tests performed. The energy consumption of the electrochemical unit individually—ELOX, the electrochemical unit and main pump together—ELOX + Pumping, and the overall prototype—Overall system—are presented excluding the equipment for temperature control. By analogy with the indicator used to assess water consumption, the energy consumption results are expressed referred to the amount of biomass produced. As shown in Table 4, in all instances, the power consumption of the electrochemical unit is below 3.5 kWh kg fish produced^{-1}—that is 9–23 % of the total energy required by the system. The total energy requirements ranged from 9.1 to 25.4 kWh kg fish produced^{-1}; thus, they were between 4 and 12 times greater than the energy consumed by the electrochemical unit. However, the observed energy use of the whole system is within the range of values reported in the literature regarding the total energy consumption of conventional RAS. More specifically, the general range varies widely between 2.9 and 81.5 kWh kg fish produced^{-1} (Song et al., 2019; Badiola et al., 2018; Badiola et al., 2017; Timmons and Ebeling, 2013). This variability depends on: (i) the scale—the production, (ii) the scope of the treatment—the treatment stages involved (i.e., whether the treatment involves denitrification or disinfection stages or not, and (iii) the farming and treatment conditions—i.e., biomass or stocking density, recirculating flow rate, etc. In this sense, Summerfelt et al. (2009) and Badiola et al. (2017) studied RAS that can be considered equivalent to ELOXIRAS®—i.e., involving a culture volume lower than 150 m^3, fully recirculated, and including disinfection stages. They reported 9.3–26.0 kWh kg fish produced^{-1}—excluding the temperature regulation

Table 4
Evaluation of energy consumption of RAS based electrochemical oxidation (ELOXIRAS®) at large scale for gilthead sea bream and sea bass farming.

Farming stage	Stocking conditions	Avg. recirculation rate (h^{-1})	Energy consumption					
			Referred to biomass (kWh·kg fish produced^{-1})			Referred to treated water (kWh·m^{-3})		
			ELOX Cell	ELOX Cell & Pumping	Overall	ELOX Cell	ELOX Cell & Pumping	Overall
Pre-growing	Standard	1.4	3.4	12.7	23.6	0.032	0.12	0.22
	High production	1.1	1.8	5.0	9.1	0.068	0.18	0.32
On-growing	Standard	1.1	2.2	11.6	25.4	0.022	0.12	0.25
	High production	0.9	3.2	7.44	13.7	0.098	0.23	0.41

equipment as well, when applicable. Based on that, the energy performance of the RAS based on electrochemical oxidation is regarded as comparable to that of those RAS based on conventional technology.

With respect to the influence of stocking in the energy use, no clear trend is observed when the energy consumption is analysed referred to the amount of biomass produced. This is because the amount of biomass produced depends to a big extent on the variables involved in fish farming, e.g., fish size, feeding (through the *SFR*), etc., which are not necessarily related to the water treatment. For that reason, other variables are preferable for standardization purposes. In this sense, energy consumption is also expressed in Table 4 referred to the volume of water treated per unit of time. As observed, the energy consumed by the electrochemical treatment for the tests conducted under high production stocking conditions is significantly higher than that obtained for the tests under standard stocking conditions—0.068 and 0.098 $kWh \cdot m^{-3}$ (high production), instead of 0.032 and 0.022 $kWh \cdot m^{-3}$ (standard production) for pre-growing and on-growing tests, respectively. This is mainly due to two main reasons: (i) the generation of pollutants is proportional to the biomass density, and (ii) the recirculation rate—related to the treatment flowrate—applied in the tests conducted under high production stocking conditions was lower than that of their standard counterparts, leading to an increase in the concentration of the pollutants in water. In summary, when working with high biomass densities, it is necessary to supply more energy to the electrochemical unit to remove the pollutants from the rearing water. However, this increase is mitigated by the low contribution of the electrochemical treatment to the total energy consumption of the system. Values of 0.22–0.25 $kWh \cdot m^{-3}$ were obtained for standard stocking conditions, while slightly higher values of 0.32–0.41 $kWh \cdot m^{-3}$ were obtained for high production stocking conditions, with a biomass density almost twice the regular; for that reason, a slight increase in the energy requirements is expectable.

4. Conclusions

In this work, electrochemical oxidation has been studied as an innovative alternative to conventional biofiltration for water treatment in marine RAS, through the simultaneous farming of gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*) in a real environment. The study consisted in four tests—5–25 weeks—involving two farming stages and stocking conditions. Treatment intensity was adjusted as required during the execution of the tests.

The electrochemical treatment performance was tested in terms of TAN and bacteria removal, showing a high pollutant removal capacity and disinfection potential that led to an adequate water quality for farming. More specifically, TAN removal efficacies higher than 90 % were achieved by adjusting the operating conditions. In addition, up to 3 log $CFU \cdot mL^{-1}$ reduction was ensured, contributing to the stabilization of the microbiota in the fish tanks. For that reason, at technical level, electrochemical oxidation is considered a highly-effective and competitive alternative to biofiltration.

Regarding the impact on fish production, typical indicators of growth and feeding were considered—*SGR*, *FCR*, and *SFR*. A satisfactory development of fish was observed, with adequate growth rates, relatively good use of feed, and a remarkable fish quality—endorsed by histopathological studies, even when dealing with high biomass densities. In this sense, by increasing the biomass density, electrochemical oxidation allows to increase fish production in marine RAS up to 90 % depending on the fish species, growth stage and farming conditions, thus, offering flexibility to changeable productivity requirements upon volatile market demand.

Finally, the environmental performance was assessed by means of water and energy consumption. In this regard, the RAS based on electrochemical oxidation reached the category of intensive RAS with 200–300 litters of make-up water per kg of fish produced—at least under standard production conditions; up to 50 % reduction compared to conventional technology, with the consequent benefits in water abstraction and wastewater discharge. As for the energy use, the energy requirements of the electrochemical unit represented just a small fraction of the global energy consumption in the RAS—10–25 % approx. These results suggest that electrochemical oxidation contributes to the intensification of marine RAS.

In summary, the high TAN removal rate and disinfection that electrochemical oxidation offers ensures the quality of marine RAS waters, which, ultimately, leads to a good development of fish, both in terms of health and growth. Additionally, the results of this work corroborate that electrochemical oxidation either enables fish production with reduced water consumption or allows to increase fish production without a significant rise in resource consumption. Either way, the electrochemical remediation of marine RAS waters may contribute to improve the environmental profile of the aquaculture sector, helping to meet the growing global food demand more sustainably. For that reason, electrochemical oxidation arises as a promising alternative and an innovative resource-efficient water treatment technology in marine RAS for fish production.

CRediT authorship contribution statement

Germán Santos: Validation, Investigation, Writing – original draft, Writing – review & editing, Data curation, Visualization. **Isabel Ortiz-Gándara:** Validation, Investigation, Data curation, Visualization. **Andrés Del Castillo:** Validation, Investigation, Data curation, Visualization. **Axel Arruti:** Methodology, Validation, Investigation, Supervision, Funding acquisition. **Pedro Gómez:** Conceptualization, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Raquel Ibáñez:** Conceptualization, Writing – review & editing. **Ane Urtiaga:** Conceptualization, Methodology, Writing – review & editing. **Inmaculada Ortiz:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157368.

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