A corrosion evaluation of mild carbon steel in reclaimed refinery stripped sour water

Pedro D.A. Bastos, Alexandre C. Bastos, Mário G.S. Ferreira, Maria António Santos, Pedro J. Carvalho, João G. Crespo

ABSTRACT

Reclaiming water for cooling systems in oil refineries has been strongly encouraged over the past years for decreasing the large consumption of fresh water, thus contributing to the efficient use of this valuable resource. In a recent study [Journal of Environmental Management 261 (2020) 110229], some of the authors studied the retention of phenols in refinery wastewater through reverse osmosis (RO) and found rejections of up to 98% of phenols and 99% of both chemical oxygen demand (COD) and total organic carbon (TOC). The permeates complied with the quality standards for make-up water in cooling processes. A missing aspect, important for the water to be used in the oil and gas industry, was the level of corrosivity of the new permeates. In this work the corrosion of mild carbon steel in the permeates and in the original cooling tower make-up water was studied by electrochemical techniques. The corrosion rate of steel in the permeates in aerated conditions was lower (between 0.053 ± 0.006 and 0.123 ± 0.011 mm year⁻¹) than in the make-up water (0.167 ± 0.030 mm year⁻¹), confirming their suitability for replacing make-up water in the cooling towers. The low corrosion of carbon steel was attributed to the low conductivity and absence of oxidizing species in the fluids, compared to fresh water.

1. Introduction

Oil refineries are known examples of water-intensive industries, relying on the ready availability of large quantities of water. As the water demand in these industries is constant all year-round, compared to other activities such as agricultural irrigation, opportunities arise for reclaiming water to mitigate the necessity of fresh water, thus reducing costs and lowering the environmental impact. For the past two decades industries have been subjected to environmental and legislative pressures regarding wastewater discharges, by setting a long-term goal of completely abolishing waste discharges to natural water bodies, a concept known as zero liquid discharge (ZLD) (Asano et al., 2007). The challenges associated with ZLD goals, together with the high costs of water and effluent discharge, have stimulated the reuse of wastewater. New environmental and economically viable options, and internal water recycling practices have been gradually adopted (Reis et al., 2018). Oil and gas industries have the potential to significantly reduce the load of freshwater supplies by implementing wastewater treatment and reuse technologies (Riley et al., 2018). In petroleum refineries the volume of wastewater has been reported to be 0.4–1.6 times the amount of processed crude oil (Diya’uddeen et al., 2011). Cooling water is one of the largest water demanding units in the petroleum industry and a clear example of where reclaimed water can be directly used if the appropriate operational conditions are achieved. Controlling the accumulation of dissolved minerals and organic compounds constitutes a major water quality problem for an appropriate cooling water system operation. Since a portion of the water evaporates during the cooling process, an increase in concentration of total dissolved solids (TDS) occurs and the addition of fresh auxiliary water (known as make-up water) is required. In order to remove higher TDS water, a blowdown is also applied. Therefore, using reclaimed water for direct use as make-up water in cooling towers appears as a viable possibility to reduce fresh water costs and overcome water depletion.

Wastewater generated from crude oil refining processes incorporates an assortment of pollutants such as oil and grease, phenols, sulphides and ammonia. Regarding environmental impact, phenols are amongst...
the major hazardous compounds in industrial wastewater due to their low biodegradability, toxicity and ecological effects on the aquatic environment as well as remaining in the environment for long periods of time and rapidly contributing to a high chemical oxygen demand (COD) (Mohammadi et al., 2015; Villegas et al., 2016). The presence of a heavy organic load of phenols in refinery stripped sour water often blocks the possibility for internal reuse. Moreover, even residual amounts of sulphides and ammonia (often < 1 and <30 ppm, respectively) (Addington et al., 2011) may pose threats to water quality regarding corrosion and biofouling if reused in cooling processes. Therefore, a treatment technology that allows the retention of these pollutants, while generating water for direct reclamation, is required for the internal reuse of stripped sour water.

Among the technologies for phenol removal from wastewaters that have been reviewed over the past few years (Mohammadi et al., 2015; Villegas et al., 2016), membrane separation technologies have been given particular attention due to low energy consumption, low operating cost, and easy scale up by membrane modules. Reverse Osmosis (RO), for instance, has been widely used at water reclamation plants where high purity water is indispensable, such as in boiler feedwater systems, with some systems designed to remove 99% of the dissolved minerals and provide high-quality water to the boiler (Judd, 2003). Reverse osmosis processes have also been recently studied as a stand-alone technology in water treatment (Albergamo et al., 2020), avoiding capital costs associated with pre-treatment steps, if the appropriate conditions are met.

In a recent study (Bastos et al., 2020) membrane filtration through RO was applied over a 6-month period to stripped sour water from the cracking complex of the largest Portuguese oil refinery, located in Sines. The resulting permeates showed a reduction of up to 98% of phenolic content and 99% of both chemical oxygen demand (COD) and total organic carbon (TOC), therefore complying with the water quality standards for cooling processes. However, a complete wastewater reclamation study should involve not only the compliance of chemical and physical parameters to the standards, but also the corrosivity of the fluid towards the materials it will contact in future reuse (Dario et al., 2009; Lee et al., 2018). Corrosion is, in fact, a common problem in our metals-based society, with costs ranging 3–5% of gross domestic product (GDP) every year in each country (Koch et al., 2002; Kruger, 2011). Preventing corrosion, as well as scaling and biofouling, are key aspects in cooling systems, and a requisite for any new technology for reclaiming water (Asano et al., 2007). The presence in reclaimed water of aggressive ions such as chlorides, sulphides and sulphates facilitates the corrosion process, either uniform or localized (pitting, crevice or stress corrosion cracking), depending on water parameters like pH, conductivity, alkalinity and dissolved oxygen.

The present work complements the previous study on membrane filtration wastewater treatment (Bastos et al., 2020) by comparing the corrosion of the carbon steel used in the cooling towers of the Sines refinery when immersed in the RO permeates or in the original fresh make-up water, therefore providing a complete evaluation of the wastewater reclamation at the oil refinery. The crude oils processed in the refinery can vary significantly and so varies the composition of the refinery wastewater. However, the studied RO filtration process was able to produce permeates with close compositions, despite the variations in the feed wastewater. In any case, for this work three permeates were collected over a 6-month period, from winter to spring, in such a way that they could fully comprehend the limits of the wastewater composition oscillations that were registered during 2 years of refinery operation and wastewater sampling and characterization. Hence, it was possible to warrant the representativeness of the permeate solutions used for the corrosion testing.

## 2. Materials and methods

### 2.1. Refinery wastewater, make-up water and chemical analyses

Refinery wastewater samples were directly withdrawn from the effluent of the sour water stripping unit of the cracking complex at the Sines refinery. Three samples of refinery wastewater were collected from the sour water stripping unit over a 6-month period and were fed to the RO membrane unit, filtered in concentration mode up to a volumetric concentration factor of 3. All tests were conducted at an operating pressure of 50 ± 1 bar and 22 ± 2 °C. Samples from the initial wastewater and permeates (P1, P2, P3) obtained after reverse osmosis were analysed by standard methods for oil and grease and phenols concentration (SMEWW 5520 C/F and SMEWW 5530D, respectively). Chemical oxygen demand (COD) was analysed by the LCI 500 cuvette kit (Hach Lange, GmbH, Germany) in agreement with ISO 15705 and total organic carbon (TOC) was determined through a total carbon analyser TOC-VCSH (Shimadzu, Japan). Ammonia and phosphates content were measured by a San++ Continuous Flow Analyzer (Skalar, The Netherlands). Turbidity values were acquired through a portable turbidity meter (Hanna Instruments, USA). Total dissolved solids (TDS), pH and conductivity were measured with a Mettler Toledo Seven Multi Equipment with Inlab Expert Pro and Inlab 731 electrodes. A sample of make-up water was collected from the refinery cooling tower and the pH, TDS, conductivity, turbidity and COD were analysed through the same methods referred above.

### 2.2. Corrosion measurements

The corrosivity of the original refinery make-up water (MW) and of the permeates P1, P2 and P3 towards mild carbon steel was assessed by electrochemical methods. In addition, to simulate the case where the cooling fluid is a mixture of the reclaimed water and MW, each permeate P1, P2 and P3, was mixed with MW in a 50/50 (wt%) proportion, giving origin to mixtures Mix 1, Mix 2 and Mix 3. Mild carbon steel pieces of 1 × 1 cm² were electrically connected to a copper wire and embedded in epoxy resin. The surface of the mounts was abraded with SiC paper down to 1200 grit, rinsed in distilled water, followed by ethanol. The measurements were performed with a PGSTAT302N Potentiostat (Autolab Metrohm), in electrochemical cells with a three-electrode arrangement, consisting of the working electrode (resin mounted steel sample), a platinum counter electrode and a saturated calomel electrode (SCE) as reference. Polarization curves were measured after 1 h of immersion either in aerated conditions or after bubbling argon for 30 min to remove dissolved oxygen. Separate anodic and cathodic sweeps were obtained with independent samples at a scan rate of 1 mV s⁻¹. Polarization resistance measurements were performed from −15 mV to +15 mV versus the corrosion potential with a scan rate of 1 mV s⁻¹. Four replicate measurements were acquired for each condition.

### 3. Results and discussion

#### 3.1. Reverse osmosis tests and permeates characterisation

The analytical data obtained for the wastewater samples, as well as the final permeates, are presented in Table 1 and compared against quality standards for reused water and with the make-up water from the Sines Refinery cooling tower. All permeates were in line with the quality standards for water reuse, regarding the most relevant parameters in water quality management, including pH, conductivity and TDS. The COD values are below the specifications, which leads to the assumption that the remaining vestigial organic matter will not compromise water quality upon reuse. Moreover, scaling or biofouling phenomena are not likely to occur, due to lack of salts or nutrients for microbial growth, such as phosphate-based compounds. Evidently, the make-up water also
lies within the water quality standards regarding all parameters studied, since the facility resorts to industrial water for cooling processes.

3.2. Corrosion tests

Since the RO permeates complied with the specifications for reuse as make-up water in cooling processes, a corrosion evaluation was performed to verify their compatibility with the carbon steel used in the cooling towers of the Sines refinery. The make-up water currently in use was also tested and served as control.

Fig. 1 presents polarization curves measured after 1 h of immersion. The cathodic sweeps (Fig. 1a) reveal the response of species in the environment that can support the oxidation of steel. The reduction of dissolved oxygen dominates the curves from the corrosion potential ($E_{\text{corr}}$) down to about $0.9 \ V_{\text{SCE}}$ and then the reduction of water becomes dominant for more negative potentials. The distinct shapes of these two regions are more evident in the curve of MW. The anodic sweeps show the current of steel oxidation as the potential gets more positive than $E_{\text{corr}}$. The current is smaller in the permeates than in the MW reference. This is an indication of a lower corrosion rate of the steel when exposed to the new permeate samples.

Quantitative values could have been obtained by the Tafel extrapolation method, but the abnormal inclination of the curves (due to the high ohmic drop in solution during polarization resulting from the low conductivity of the tested waters) would lead to an erroneous estimation of the corrosion rates. Consequently, the polarization resistance method was preferred because it just needs a small polarization around $E_{\text{corr}}$ and the involved currents are very small (in the μA cm$^{-2}$ range) leading to a much more reduced ohmic drop in solution. The experimental plots are shown in Fig. 2 where, for a better comparison of the different curves, the potential axes depict overpotentials ($\eta = E_{\text{applied}} - E_{\text{corr}}$) instead of $E_{\text{corr}}$. Mixtures of the permeates P1, P2 and P3, with MW in a 50/50 (wt %) proportion were also tested to simulate the case where the cooling fluid is a mixture of the reclaimed water and MW. The mixtures of permeates and make-up water were prepared and studied considering that treating the stripped sour water using reverse osmosis on an industrial scale could generate a permeate flow rate that would meet about half of the make-up water demand of one of the refinery cooling towers. Therefore, a study of the corrosion evaluation using the prepared mixtures was considered relevant for a realistic assessment of the final water

### Table 1

Analytical data for feed and permeate characterization from cross-flow filtration studies and water reuse quality standards.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Feed</th>
<th>Permeate (P1)</th>
<th>Feed</th>
<th>Permeate (P2)</th>
<th>Feed</th>
<th>Permeate (P3)</th>
<th>Water reuse quality standards$^a$</th>
<th>Sines Refinery cooling tower</th>
</tr>
</thead>
<tbody>
<tr>
<td>$pH$</td>
<td>8.83</td>
<td>6.61</td>
<td>9.35</td>
<td>6.84</td>
<td>10.41</td>
<td>7.79</td>
<td>6.5–9.0</td>
<td>7.81</td>
</tr>
<tr>
<td>Conductivity ($\mu S \ cm^{-1}$)</td>
<td>222</td>
<td>7.77</td>
<td>86.3</td>
<td>8.82</td>
<td>335</td>
<td>109.1</td>
<td>$^b$</td>
<td>1095</td>
</tr>
<tr>
<td>TDS (ppm)</td>
<td>71</td>
<td>3.77</td>
<td>46</td>
<td>4.59</td>
<td>101</td>
<td>54.63</td>
<td>500</td>
<td>548</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>–</td>
<td>2.21</td>
<td>16.8</td>
<td>1.02</td>
<td>14.5</td>
<td>1.18</td>
<td>1.76</td>
<td>5.34</td>
</tr>
<tr>
<td>COD (mgO$_2$/L)</td>
<td>857</td>
<td>29.5</td>
<td>1605</td>
<td>9.5</td>
<td>1577</td>
<td>54.7</td>
<td>75</td>
<td>5.34</td>
</tr>
<tr>
<td>TOC (ppm)</td>
<td>273.3</td>
<td>8.48</td>
<td>399.4</td>
<td>3.72</td>
<td>664.7</td>
<td>13.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Phenols (ppm)</td>
<td>160</td>
<td>4.4</td>
<td>128</td>
<td>3.2</td>
<td>184</td>
<td>6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total O&amp;G (ppm)</td>
<td>4</td>
<td>&lt;LOQ</td>
<td>771</td>
<td>&lt;LOQ</td>
<td>98</td>
<td>&lt;1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ammonia (ppm)</td>
<td>13.62</td>
<td>0.58</td>
<td>9.47</td>
<td>0.65</td>
<td>37.84</td>
<td>9.45</td>
<td>$^b$</td>
<td>548</td>
</tr>
<tr>
<td>Phosphates (ppm)</td>
<td>&lt;LOQ</td>
<td>&lt;LOQ</td>
<td>&lt;LOQ</td>
<td>&lt;LOQ</td>
<td>&lt;LOQ</td>
<td>&lt;LOQ</td>
<td>$^b$</td>
<td></td>
</tr>
</tbody>
</table>

LOQ – limit of quantification.

$^a$ Asano et al.(2007).

$^b$ Accepted as received.

![Fig. 1.](image-url)  
**Fig. 1.** a) Cathodic and b) anodic polarization curves of carbon steel after 1 h of immersion in make-up water (MW) and permeates P1, P2 and P3.
to be reused in the cooling towers.

Tests with the permeates and MW were repeated in deaerated conditions (after bubbling argon for 30 min) in order to determine whether oxidant species, other than oxygen from the air, exist in the samples that could cause corrosion of the steel. Note the tenfold decrease in the current density scale in plot c).

The polarization resistance values ($R_p = \Delta E_{app}/\Delta i$) are presented in Table 2 and Table 3. The corrosion current ($i_{corr}$) can be calculated from the $R_p$ values using the Stern-Geary equation (Stern and Geary, 1957),

$$i_{corr} = \frac{B}{R_p}, B = \frac{\beta_a \beta_c}{2.3 (\beta_a + \beta_c)}$$  \hspace{1cm} (1)

where $\beta_a$ and $\beta_c$ are the Tafel constants of the anodic and cathodic reactions, respectively. Since the curves in Fig. 1 are strongly affected by the ohmic drop in solution, the slopes of the Tafel lines are unreliable and, in this case, it is advisable to use a value of $B = 26$ mV (Fontana, 1987). This procedure is commonly adopted when the electro-kinetics parameters are unknown and is based on the fact that most Tafel slopes lie within well-defined limits (Fontana, 1987). Using mean values for $\beta_a$ and $\beta_c$ yields $B = 26$ mV which yields corrosion rates differing by no more than a factor of 3 from the actual rates, when applied in Eq. (1) (Fontana, 1987). More important, since the metal and solutions are similar, the reactions are the same and so are the kinetic parameters. Hence, the error in using an approximate value of $B$ will be the same for all systems and will not affect their relative rates. The corrosion current density can be converted to the units of mass loss per unit area per unit time after applying the Faraday laws of electrolysis,

$$\frac{m}{A t} = \frac{M}{n F} i_{corr}$$  \hspace{1cm} (2)

where $m$ is the mass lost during time $t$ in a surface area $A$, $M$ is the molar mass of iron, $n$ is 2 (for the reaction Fe → Fe$^{2+} + 2e^-$), $F$ is the Faraday constant (96,485 C mol$^{-1}$) and $i_{corr}$ is the corrosion current density determined in Eq. (1). The direct application of Eq. (2) gives the mass loss per second in 1 cm$^2$. Dividing by the iron density gives the corrosion rate in the form of thickness reduction. The corrosion rates in the three different units are presented in Tables 2 and 3.

The corrosion rates were higher for the MW than for the permeates which confirms that the RO process yields water with lower corrosivity. The reason is attributed to the lower conductivity of the permeates resulting from the purification process. The higher resistive medium increases the global resistance of the corrosion cell and decreases the corrosion current by introducing a fraction of resistance control to the polarization (Revie and Uhlig, 2008). Hilbert has discussed the importance of low conductivity water for the low corrosion of carbon steel piping in district heating systems (Hilbert, 2006). Lee et al. compared the effects of tap water and reclaimed water on the corrosion of the inner surface of pipes and observed that the reclaimed water with higher ionic content showed higher corrosion rate than tap water (Lee et al., 2018). As the permeates are mixed with MW, the conductivity increases and the corrosion rates become closer to that of the original MW.

Another important result is that upon removing the dissolved oxygen

**Table 2**

<table>
<thead>
<tr>
<th>E$_{corr}$ (V vs. SCE)</th>
<th>$R_p$ (Ω cm$^2$)</th>
<th>Corrosion rates</th>
<th>Mass loss (mg cm$^{-2}$ y$^{-1}$)</th>
<th>Thickness reduction (mm y$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Make-up water (MW)</td>
<td>-0.677 ± 0.043</td>
<td>1802 ± 235</td>
<td>14.4 ± 2.6</td>
<td>131.3 ± 23.4</td>
</tr>
<tr>
<td>Permeate 1 (P1)</td>
<td>-0.697 ± 0.036</td>
<td>4927 ± 650</td>
<td>5.28 ± 0.70</td>
<td>480.0 ± 6.3</td>
</tr>
<tr>
<td>Permeate 2 (P2)</td>
<td>-0.610 ± 0.043</td>
<td>5662 ± 812</td>
<td>4.59 ± 0.66</td>
<td>41.8 ± 6.0</td>
</tr>
<tr>
<td>Permeate 3 (P3)</td>
<td>-0.661 ± 0.051</td>
<td>2453 ± 211</td>
<td>10.6 ± 0.90</td>
<td>96.5 ± 8.3</td>
</tr>
<tr>
<td>Mix 1</td>
<td>-0.719 ± 0.033</td>
<td>2388 ± 722</td>
<td>11.7 ± 3.5</td>
<td>106.6 ± 31.9</td>
</tr>
<tr>
<td>Mix 2</td>
<td>-0.637 ± 0.039</td>
<td>2441 ± 245</td>
<td>10.7 ± 1.1</td>
<td>96.9 ± 9.7</td>
</tr>
<tr>
<td>Mix 3</td>
<td>-0.734 ± 0.056</td>
<td>1880 ± 263</td>
<td>14.0 ± 1.9</td>
<td>127.9 ± 17.0</td>
</tr>
</tbody>
</table>

**Table 3**

<table>
<thead>
<tr>
<th>E$_{corr}$ (V vs. SCE)</th>
<th>$R_p$ (Ω cm$^2$)</th>
<th>Corrosion rates</th>
<th>Mass loss (mg cm$^{-2}$ y$^{-1}$)</th>
<th>Thickness reduction (mm y$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Make-up water (MW)</td>
<td>-0.778 ± 0.032</td>
<td>22,392 ± 4980</td>
<td>1.16 ± 0.26</td>
<td>10.61 ± 2.36</td>
</tr>
<tr>
<td>Permeate 1 (P1)</td>
<td>-0.734 ± 0.044</td>
<td>35,477 ± 6829</td>
<td>0.73 ± 0.14</td>
<td>6.70 ± 1.29</td>
</tr>
<tr>
<td>Permeate 2 (P2)</td>
<td>-0.727 ± 0.039</td>
<td>42,387 ± 10,370</td>
<td>0.61 ± 0.15</td>
<td>5.60 ± 1.37</td>
</tr>
<tr>
<td>Permeate 3 (P3)</td>
<td>-0.759 ± 0.041</td>
<td>30,218 ± 6101</td>
<td>0.86 ± 0.17</td>
<td>7.86 ± 1.59</td>
</tr>
</tbody>
</table>
the corrosion rate substantially decreases, a fact expected because no
evidences of corrosion were observed with the MW in use today in the
Sines refinery. Being a closed system, no substantial oxygen is in the
fluid to feed the corrosion of steel. The importance of oxygen in the
corrosion of steel cannot be overstated. An investigation of the effect of
dissolved oxygen (DO) on internal corrosion of water pipes reported that
a reduction of DO concentration from 9 ± 0.5 mg/L to 2 ± 0.5 mg/L
leads to a 75% decrease of the corrosion rate of carbon steel (from 133 to
32 mg cm⁻² y⁻¹) and concluded that DO could be used as a major
parameter in controlling the corrosion of water pipes (Jung et al., 2009).
In another study, steel coupons after 1 day immersed in aerated and
daerated synthetic RO platform water (215 ppm Cl⁻, 18 ppm SO₂⁻, 148 ppm Na⁺, pH = 5.65, conductivity = 766 μS cm⁻¹) were corroding
at 200 μm y⁻¹ (with oxygen), while in the same solution deaerated with
ultra-high purity nitrogen (99.999%) the corrosion rate was only 5 μm
y⁻¹, 40× slower (Fredj et al., 2012).

It is also noticeable that, with the exception of oxygen from the air, no
oxidant agents are present in the permeates that could cause significant
damage, as their corrosion rates also greatly decreased and showed a
response similar to MW, upon removal of oxygen. These results show
that the trace contaminants remaining in the permeates have negligible
effect on the corrosion of steel. It is important to restate that the
permeates used in this study represent effluents from RO purification of the
most compositionally diverse wastewaters produced in a two-year period in the refinery operation and wastewater sampling and
characterization.

As a final conclusion of these experiments, the corrosion of steel in
contact with the permeates was lower than with the MW, confirming the suitability and safe use of the permeates in the cooling system of the
refinery.

4. Conclusions

This study evaluated the corrosion of mild carbon steel in the permeates
from the reverse osmosis filtration of striped phenolic water
water to determine the feasibility of their use as make-up water in
cooling towers. The corrosion of steel in the permeates was compared
against the corrosion of steel in fresh industrial water used daily at the
refinery. The linear polarization resistance method showed that, in
aerated conditions, the corrosion rate of steel in the permeates varied
between 0.053 ± 0.006 and 0.123 ± 0.011 mm y⁻¹ while 0.167 ± 0.030
mm y⁻¹ was obtained for the make-up water. These rates are largely
controlled by the low conductivity of the permeates and the absence of
oxidizing species in the fluids, apart from oxygen. The lower corrosion
rate found in all permeates confirms their adequacy and safe use in the
cooling system of the refinery. These findings, together with the chemical
composition of the permeates compared with the feed water and the
make-up water, lead to the final conclusion that reverse osmosis showed to
be a suitable stand-alone technology for generating water from
phenolic refinery wastewater for direct use as make-up water in cooling
towers.

Declaration of competing interest

There are no conflicts of interest to declare.

CRediT authorship contribution statement

Pedro D.A. Bastos: Investigation, Visualization, Writing - original
draft. Alexandre C. Bastos: Methodology, Supervision, Investigation,
Visualization, Writing - review & editing. Mário G.S. Ferreira: Supervision, Resources. Maria António Santos: Conceptualization, Re-
sources, Supervision. Pedro J. Carvalho: Writing - review & editing, Supervision. João G. Crespo: Conceptualization, Methodology,
Supervision, Writing - review & editing.

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