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Recycled wastewater as a potential source of microplastics in irrigated soils from an arid-insular territory (Fuerteventura, Spain)

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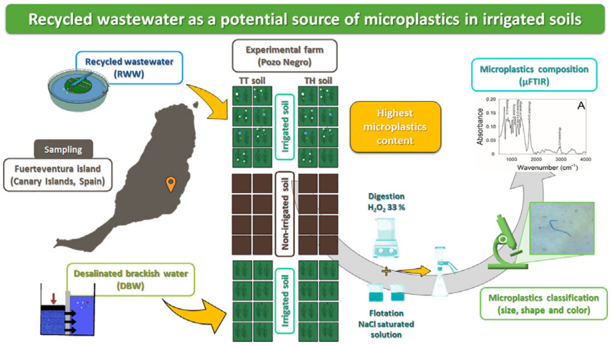
HIGHLIGHTS

- Microplastics occurrence in irrigation waters and soils irrigated with them was accomplished.
- Prevalence of cellulosic and polyester blue and transparent microfibers was found.
- Similar microplastics patterns (shapes, colors and composition) were found between waters and soils.
- Desalinated brackish water and the soil irrigated with it contained less microplastics.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, the occurrence of microplastics (MPs) in irrigation recycled wastewaters (RWWs) and a desalinated brackish water (DBW) from the arid territory of Fuerteventura (Canary Islands, Spain) was studied. Besides, the presence of MPs in two types of soils (sandy-loam and clay-loamy; with no mulch film or fertilization with sewage sludge applied) irrigated with both water qualities was addressed. Results showed the prevalence presence of cellulosic and polyester microfibers (between 84.4 and 100%) of blue and transparent colors (up to 55.6 and 33.3%, respectively), with an average length of 786.9 ± 812.1 μm in the water samples. DBW had the lowest MP concentration (2.0 ± 2.0 items·L^{-1}) while RWW showed concentrations up to 40.0 ± 19.0 items·L^{-1}. Similarities were also observed between the MPs types and sizes found in both soils top layer (0–5 cm), with an average concentration three times greater in soil irrigated with RWW than in soil under DBW irrigation (159 ± 338 vs. 46 ± 92 items·kg^{-1}, respectively). In addition, no MPs were extracted from non-irrigated/non-cultivated soils, suggesting agricultural activities as the unique source of MPs in soils of this arid area. Results show that RWWs constitute a potential source of MPs in irrigated soils that should be considered among other pros and cons linked to the use of this water quality in agricultural arid lands.

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1. Introduction

Plastic pollution is currently one of the most important environmental problems that humanity has to face. The exponential growth of plastic production since 1950s (up to 368 million of tons were produced in 2019; Plastics Europe, 2020) and the massive use of plastics, together with an insufficient/inadequate waste management/disposal, are the main causes of the global presence of plastics in every environmental compartment.

Current research is showing that one of the main concerns about plastics, apart from the fact that they remain in the environment for an extremely long time, is their constant fragmentation into ever smaller pieces called microplastics (MPs, 1 μm–5 mm) or nanoplastics (< 1 μm), depending on their final dimensions, though they are also released as such (ECHA, 2019).

Concerning the soil compartment, MP research has been hardly investigated compared to the attention paid to the marine environment (Yang et al., 2021). However, the key and numerous ecosystems services developed by soils (e.g., He et al., 2018; Wang et al., 2019; Delgado-Baquerizo et al., 2020; Ellili-Bargaoui et al., 2021), and the overall finding that MP contamination of terrestrial ecosystems might be several times higher than in oceans (de Souza Machado et al., 2018a, 2018b), has increased the attention on soil MP pollution in the last five years (Dioses-Salinas et al., 2020; Zhou et al., 2020b; Yang et al., 2021). As a result, studies are revealing that the presence and accumulation of MPs can affect several physical, chemical and biological processes, all of them extremely important for soil ecosystem functioning (de Souza Machado et al., 2018a, 2018b; Wan et al., 2019). Many of these harmful effects are not only directly linked to the MP particles themselves, but also to the possible release of toxic plastic additives (which are not linked to the polymeric matrix) or by persistent and emerging contaminants than can be retained onto their surface by different mechanisms (Nizzetto et al., 2016; Jiménez-Skrzypek et al., 2021).

In the particular case of agricultural soils, MPs presence can be related with the massive amount of plastic products used in conventional farming, for example, plastic film mulching, packaging, plastic films for greenhouse shedding, water pipes, fertilizers coatings, and substrate aeration-improving materials which eventually degrade on the field and transform into MPs (Hayes, 2019; Dioses-Salinas et al., 2020; Zhou et al., 2020a; Yang et al., 2021). However, the largest MP amounts that reach agricultural soils is through the application of sewage sludge as fertilizer (Liu et al., 2018; Corradini et al., 2019; Gao et al., 2020; Weithmann et al., 2018). Concerning irrigation water, particularly recycled wastewater (RWW), it has been studied to a much lesser extent as it also happened with organic amendments like compost and manure (Zhou et al., 2020a).

With regard to wastewater, several review articles have evaluated the detection and identification of MPs in wastewater treatment plants (WWTPs) influents and effluents, as well as the effectiveness of the treatment in their removal (Sun et al., 2019; Cheng et al., 2021; Hamidian et al., 2021; Xu et al., 2021). From such works, it is clear that WWTPs act at the same time as a sink and as a source of MPs to the environment. In the first case, they are considered a sink because wastewater contains an extremely high number of microfibers (natural, semi-synthetic and synthetic) which have been released during textiles laundering (Napper and Thompson, 2016; Yang et al., 2019) and which finally accumulate in sewage sludge (Gao et al., 2020). However, since sewage sludge is commonly used by farmers as fertilizer as a result of its high levels of organic and inorganic nutrients, its use is also a source of MPs to the agricultural system (Zubris and Richards, 2005; Gao et al., 2020; van den Berg et al., 2020). While nutrients only remain in a relatively short period in soil, plastics accumulate and persist in edaphic environments for a long time (Corradini et al., 2019; van den Berg et al., 2020). As an example, the amount of MP particles that sewage sludge delivers into European farmlands is estimated to be up to 4000 MPs granules and 670 MPs fibers per kg of dry soil in the topsoil layer (Leed and Smithson, 2019). Another important issue concerning the role of WWTPs as MPs source, is the fact that, despite the majority of MPs are eliminated during wastewater treatment (a removal rate in the range up to 90–99% has been reported in some occasions; Murphy et al., 2016; Bayo et al., 2020a, 2020b), still important amounts of them are released to the environment via wastewater discharges. As a result, they can be potentially transported to agricultural soils when RWW is used for irrigation (Bayo et al., 2020a; Prajapati et al., 2021; Ben-David et al., 2021). In this sense, and in spite of the global growing use of non-conventional water resources such as recycled urban wastewater in irrigation, particularly in arid and semiarid areas, MPs load reaching agricultural soils from this type of water have been poorly evaluated yet (Zhang and Liu, 2018; Zhou et al., 2020a).

This is the case of the work of Zhou et al. (2020a) in which several Chinese soils covering mulching and no-mulching farmlands were analyzed, as well as irrigation waters from rivers where nearby wastewater discharges occurred. Mulching soils had the higher amounts of MPs, mainly fragments of 1–3 mm length. Even though, authors found an important contribution of irrigation water, plastic wastes decomposition and compost application on the MPs contents of both groups of soils. In another work carried out by Zhang and Liu (2018), authors determined the concentration of MPs in soils from China irrigated with wastewater and treated with sewage sludge, finding MPs in all the soils (mainly fibers) at extremely high concentrations (7100–42,960 items·kg^{-1}), which could be associated with both irrigation and fertilization practices. To the best of our knowledge, only the work of Zhou et al. (2020a) has simultaneously analyzed both soils and irrigation water samples looking for a possible correlation between them. However, the high amounts of external MPs sources did not provide concluding data. Therefore, the aim of this work is to study the occurrence of MPs in soils as a result of their irrigation with RWW. Our main research hypothesis is that RWW, despite the different treatments that are currently being applied in WWTPs, could constitute a significantly greater source of MPs in irrigated soils compared to the desalinated brackish water (DBW) used as control. For this purpose, two types of agricultural soils located at Fuerteventura Island (Spain) and irrigated with RWW and DBW on a regular basis, were considered. No other possible MP source like the use of plastic mulches for soil coverage or sewage sludge as fertilizer were applied. In addition, for comparison purposes, RWW obtained from different WWTPs and also used by local farmers for irrigation, were analyzed to investigate a possible release of MPs into different farmlands.

2. Material and methods

2.1. Study area

This study was developed at the volcanic island of Fuerteventura (Canary Islands, Spain), located between 28°45′ and 28°02′ N and 13°49′ and 14°20′ W, and 115 km off the NW coast of Africa (Fig. 1). Fuerteventura Island constitutes one of the most arid territories of the European Union, with most of its surface suffering intense desertification processes (Díaz et al., 2011). The island receives on average 150 mm of rainfall per year, with no more than 250 mm in any area. The rainfall is seasonal, from October to March, with high inter-annual variability. Mean annual temperature is approximately 20 °C, average annual relative humidity ~64%, average wind speed ~3.4 m·s^{-1}, average radiation ~19 MJ·m^{-2}·day $^{-1}$, and an average of 10.6 h sunshine per day. The net combination of these climatic factors leads to a reference evapotranspiration (ET_o) of ~1700–1800 mm·year^{-1}. Due to the extreme aridity, current agricultural production mainly relies on the use of non-conventional water resources such as RWW, which availability in large volumes all over the year has exponentially increased with the tourist industry development (around 15 hm^3 of urban wastewater are treated every year) (Dorta-Santos et al., 2014). Urban wastewater is treated in several conventional different size WWTPs spread over the island territory, which final effluents are supplied to farmers for irrigation.

The soil study was conducted on “Pozo Negro” experimental farm (owned by the Cabildo Insular of Fuerteventura) located in the central-east part of the island (see Fig. 1). In this farm, several soil experimental plots have been dedicated since November 2016 to alfalfa (*Medicago sativa* L.) production to supply forage to the local market with a high demand for

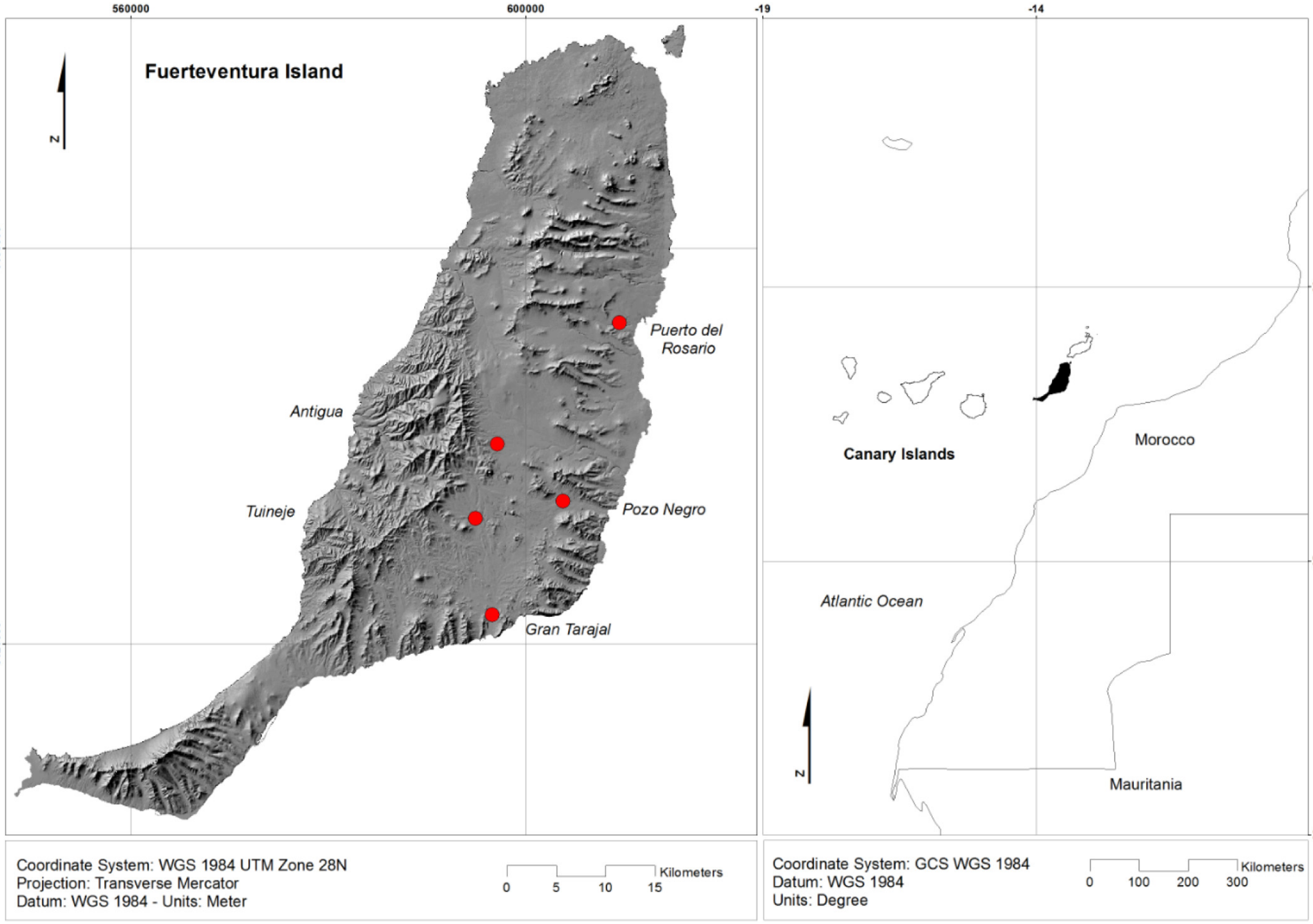

Fig. 1. Location of Pozo Negro experimental farm and the four WWTPs in Fuerteventura (Canary Islands, Spain); Puerto del Rosario (WWTP-1); Gran Tarajal (WWTP-2); Tuineje (WWTP-3); Antigua (WWTP-4); Pozo Negro (BWDP). Image obtained using ArcGIS Desktop software (ESRI's ArcGIS Desktop 10.8 software). WWTP: Wastewater treatment plant; BWDP: Brackish water desalination plant.

livestock feed, one of the main economic activities in the island. Alfalfa was grown in two different soil types (two experimental fields): Typic Torrifluvents (TT) and Typic Haplocambids (TH) (Soil Survey Staff, 2014). The TT soil was 60–80 cm thick on average, and sandy-loam in texture at the top layer (0–20 cm; clay 143 $g \cdot kg^{-1}$, silt 179 $g \cdot kg^{-1}$, sand 678 $g \cdot kg^{-1}$), whereas the TH soil was thicker (90–100 cm) and clay-loamy at 0–20 cm (clay 293 $g \cdot kg^{-1}$, silt 506 $g \cdot kg^{-1}$, sand 201 $g \cdot kg^{-1}$). Both soils presented the following characteristics/conditions: (i) they were isolated from a relatively high anthropogenic pressure due to their location (i.e., far from urban centers and high-load roads), (ii) they were used in the past for low intensity/input rainfed agricultural production, mainly cereals. Since the mid-eighties they were abandoned and without agricultural activity until 2010, when they began to be used as experimental fields for evaluation of RWW irrigation effects in soil quality. In any case, plastic mulches for soil coverage or sewage sludge as fertilizer were not applied. Fig. S1 of the Supplementary Material shows the configuration of one of the experimental fields (soil TH).

Each experimental field was divided into 24 plots (a total of 48 plots of 9 m^2 in size each): half of them (24 plots) were TT soil and the other half (24 plots) TH soil. Among the 24 plots of each soil, 8 were cultivated and irrigated with RWW, 8 were also cultivated and irrigated with DBW, while 8 were not cultivated nor irrigated. The RWW (named RWW-2) was obtained from a nearby WWTP (named WWTP-2) located at Gran Tarajal (see Fig. 1), a coastal touristic village, where the incoming urban wastewater (mainly black and grey water; Ragoobur et al., 2021) is subjected to screening, grit removal, primary settling, aeration/activated sludge and chlorination. The DBW was generated from saline groundwater drawn from a depth of 45–60 m and pretreated by filtering with filter cartridges prior to dual membrane reverse osmosis at a brackish water desalination plant (BWDP) in the same experimental farm. The experimental fields were equipped with localized automatic irrigation systems with lines spaced 0.25 m apart at the soil surface. Pressure-compensating and non-leakage drippers with delivery rates of 2.2 $L \cdot h^{-1}$ were spaced 0.25 m in the irrigation lines. The irrigation rates applied varied monthly during the crop period, adjusted to match approximately 125% of ETo, calculated from the Penman-Monteith-FAO model using data obtained from an on-site weather station. Fertilization only consisted of an initial 3–4 $kg \cdot m^{-2}$ of mature goat manure incorporated to the first 20 cm of the topsoil.

2.2. Water and soil samples collection

RWW samples were collected at four conventional WWTPs (see Fig. 1) and one BWDP which part of the final effluents is used by the farmers in the island for irrigation. The plants are located in the vicinity of Puerto del Rosario (WWTP-1), Gran Tarajal (WWTP-2), Tuineje (WWTP-3), Antigua (WWTP-4) and Pozo Negro (BWDP). Final effluents of WWTPs are obtained from black and grey water after a preliminary treatment (screening and grit removal), a primary treatment (primary settling), a secondary treatment (aeration/activated sludge) and a tertiary treatment for disinfection (chlorination). In addition WWTP-1 also uses sand filters as a tertiary treatment (see Table S1 of the Supplementary Material for more information about WWTP characteristics). A total of 60 water samples were collected in three different days (21st May 2021; 29th June 2021; and 14th July 2021; sampling days and daily time are shown in Table S1 of the Supplementary Material). In each WWTP, three glass bottles of

500 mL previously washed with Milli-Q water and heated to 550 °C were filled with the irrigation waters. The same procedure was repeated for the DBW. Two liters of the final effluents as well as for the DBW were collected in white polyethylene (PE) bottles for their physicochemical characterization. After collection, all samples were stored at 4 °C, taken to the laboratory and immediately analyzed to avoid any microbial growth.

For soil MPs assessment, one soil sample was collected approximately in the center of each of 48 selected plot (under a dripper for those plots with irrigation; Fig. 1c of the Supplementary Material) on 17th February 2021 (total samples = 48; 16 irrigated with RWW, 16 irrigated with DBW and 16 without irrigation nor crop). All samples were taken from the top soil (0–5 cm depth) using stainless-steel core samplers (20 cm^2 in diameter and 5 cm height; Fig. 1d of the Supplementary Material) previously rinsed three times with Milli–Q water, covered with clean aluminum foil and stored in the darkness until their analysis at the laboratory.

2.3. Physicochemical analysis of water and soil samples

Along with taking water samples in each WWTP and BWDP for the extraction of MPs, a water sample was collected in 2 L PE bottles and taken to the laboratory for physicochemical characterization. The analyses performed followed the Standard Methods for the Examination of Water and Wastewater (APHA, 1998). The following parameters were assessed: pH, electrical conductivity (EC), cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+), anions (Cl^-, SO_4^{2-}, PO_4^{3-}, NO_3^-), total nitrogen (TN), total phosphorous (TP), chemical oxygen demand (COD), biological oxygen demand (BOD), total suspended solids (TSS), and turbidity.

After weighting all the soil contained in each core, which was mixed and homogenized in a glass beaker, a soil subsample (~ 10 g) from each core was used to determine water content by weighing it accurately on an analytical balance and drying it at 105 °C during 24 h (3 replicates per sample). Humidity percentage allowed the calculation of soil bulk density from fresh weight of the entire core. Another soil subsample from each core was air-dried and passed through a 2 mm sieve for subsequent analysis. The assessed parameters were pH ($pH_{1:5}$) and electrical conductivity in 1:5 water extract ($EC_{1:5}$), and oxidizable soil organic carbon (SOC). All the soil analyses followed Standard Methods (Soil Survey Staff, 1996).

2.4. Microplastics extraction from water and soil samples

Once at the laboratory, the exact volume of the water samples was measured, and they were vacuum filtered through glass fiber filters of 0.45 μm (VWR International) inside a glove box. The filtrates were quickly introduced in glass Petri dishes and visualized under a trinocular light stereomicroscope with magnifications ×0.65 – ×5.5 (Euromex Nexius Zoom EVO, The Netherlands) and with an image analysis system (Levenhuk M1400 PLUS - 14Mpx digital camera with the Levenhuk Lite software) which was used to measure MPs dimensions.

Concerning soil samples, 10 g of the mixed soil from each core were accurately weighted in an analytical balance and digested during 2 h with 40 mL of 33% (*w*/*v*) H_2O_2 at 60 °C in order to remove the organic matter (constant stirring at 300 rpm). Afterwards, 100 mL of a NaCl saturated solution (approximate density of 1.2 $g{\cdot}cm^{-3}$) were added and, after stirring for 1 min, the solution was left to decant for 1.5 h and filtrated under vacuum through a 50 μm stainless-steel filter (Labopolis SL, Madrid, Spain) previously washed with Milli-Q water obtained from a Milli-Q Gradient A10 system from Millipore (Burlington, MA, USA). The flotation procedure was repeated four times. The filters, which were immediately introduced in Petri dishes, were visualized under the stereomicroscope to identify and classify plastic particles according to their shape, size and color. The lower length limit of the particles studied was ~50 μm and the viewing time per filter was between 1 and 1.5 h. To visually establish if a particle is made of plastic, the criteria of Hidalgo-Ruz et al. (2012) and Marine & Environmental Research Institute (2017) were met, even though, a subset of samples was confirmed by microFourier Transform Infrared Spectroscopy (μFTIR). A diagram with the analytical procedure followed for the extraction of MPs in irrigation water and soil samples is shown in Fig. S2 of the Supplementary Material.

2.5. MicroFTIR analysis

The chemical composition of a randomly distributed subsample of microparticles (~ 100% and 16.7% of MPs found in soils and waters, respectively), which included fibers of each filter in each area, was analyzed by μFTIR using a Perkin-Elmer Spotlight™ 200 Spectrum Two instrument with a mercury cadmium telluride detector. Each microparticle was placed on a potassium bromide slide, and its spectrum was recorded in microtransmission mode, considering the following detection parameters: spot of 50 μm, 32 scans, and spectral range of 550–4000 cm^{-1}. All particles' spectra were compared with Omnic 9.1.26 database (ThermoFisher Scientific Inc., Massachusetts, USA) and with those spectra from our own database. Microparticles were considered as plastics when the match confidence was >70%. Polyethylene terephthalate (PET) was classified as "polyester (PES)" since it is a thermoplastic polymer resin of the polyester. Natural fibers (cellulose, cotton and linen) and semi-synthetic fibers (rayon/viscose/cellophane, lyocell/Tencel) as well as both cotton and linen with non-natural colors that consists of cellulose, were classified as cellulosic since their spectra are practically identical and, therefore, they are difficult to differentiate especially in the case of the microparticles found in the environment due to weathering processes.

2.6. Contamination control

All material used was plastic-free. Nonvolumetric glassware was cleaned by heating up to 550 °C for 4 h in a muffle Carbolite CWF 11/13 instrument, while volumetric glassware was cleaned using a NoChromix® solution from Godax Laboratories (Cabin John, MD) in sulfuric acid (95% *w*/*w*, VWR International) for 24 h. They were all covered with aluminum foil. Before their use, all laboratory materials were washed with Milli-Q water and previously filtered through a polyvinylidene fluoride (PVDF) 0.22 μm filter. Milli-Q water was also used to prepare the NaCl saturated solution. Both H_2O_2 33% (*w*/*v*) and NaCl saturated solutions were also filtered through a 0.22 μm filters of PVDF.

Laboratory controls (full sample pretreatment without soil) were also analyzed with every batch of samples in order to check that no laboratory contamination took place. Additionally, checks for contamination during sampling and sample processing were made by exposing filters to the air of the laboratory, whenever samples were open to the laboratory environment. In general, special care was taken to minimize airborne MPs contamination, which included the use of a glove box. Besides, orange laboratory coats frequently treated with a lint remover were also used to quickly detect possible laboratory contamination, since no orange MPs are frequently found in these types of samples.

2.7. Statistical analysis

Statistical methods were implemented using Statistical Package for the Social Sciences (SPSS, Version 26.0). The level of significance for all tests was set to $p < 0.05$. To detect differences in MPs debris (items per liter or items per kg) and MPs length (μm), among irrigation water qualities and between soils under different treatments an ANOVA and post-hoc Tukey's test were used. A Kuskal–Wallis test and a non-parametric Tukey-type multiple comparisons test were used when parameters did not conform to a normal distribution (Kolmogorov Smirnov test) and homogeneity of variance (Levene test).

3. Results and discussion

3.1. Physicochemical characterization of water and soil samples

Table S2 of the Supplementary Material shows the physicochemical characterization of the irrigation waters sampled during the study period.

As expected, TSS were generally higher in RWWs than in DBW (24.0 vs. 0.5 mg·L^{-1}, respectively), although a great variability can be observed in this and most of the assessed parameters between RWWs from different WWTPs. All water qualities had a similar pH (~ 7.1), but EC values increased significantly in RWWs waters with regards to DBW (1.9 vs. 0.4 dS·m^{-1}, respectively). That EC increased was mainly linked to a much higher concentration in Na$^+$ and Cl$^-$. Therefore, in RWWs levels of Na$^+$ and Cl$^-$ reached up to 600 mg·L^{-1} in some samples. Concentration of TN and TP were also significantly larger in RWWs than in DBW (i.e., TN and TP levels were approximately 6.5 and 7.2 times greater, respectively, in RWWs than in DBW). Similarly, COD and BOD values in RWWs significantly exceeded those in DBW; for example, COD and BOD reached average values of 73 and 22 mg·L^{-1}, respectively, in RWW-3, while in DBW from Pozo Negro averaged 2 and < 1 mg·L^{-1}, respectively). These data indicate that, in general terms, RWWs may be considered a potential source of organic matter and essential nutrients for plants and microorganisms. Notably, RWW samples with a higher amount of organic material were those from smaller size treatment plants (i.e., WWTP-3 and WWTP-4).

Table S3 of the Supplementary Material displays the main characteristics of both types of soil under different irrigation treatments. Water content was slightly higher in soil TH compared to soil TT, and as expected, significantly larger in irrigated soils with regards to rainfed soils. Bulk density ranged between 1.0 and 1.6 g·cm^{-3}, being only in soil TT significantly higher in non-irrigated/non-cultivated soils. The reaction of both soils was alkaline (pH > 8.0), and significantly greater in non-irrigated/non-cultivated soils. Lowest salinity levels were found in DBW irrigated soils accordingly with the very low EC in DBW (Table S2 of the Supplementary Material). Control soils showed a very small organic carbon concentration (< 5 g·kg^{-1}) as usual in soil of arid areas. These concentrations were significantly high (particularly in TH soil) under crop and irrigation with both types of water, with averages slightly superior under RWW irrigation.

3.2. Occurrence of microplastics in irrigation water samples

Despite the fact that a good number of works in the literature have previously digested wastewaters with different oxidizing agents (Estahbanati and Fahrenfeld, 2016; Mason et al., 2016; Ziajahromi et al., 2017; Edo et al., 2020), in our case the final effluents were relatively clean and could be directly filtrated and visualized, though in some cases (i.e. in samples RWW-4 and RWW-3), 4–5 filters had to be used for a better visualization.

Table 1 compiles the content and classification of the MPs found it the five types of irrigation water, which includes their concentration, shape, color, length and composition, while Fig. 2 shows the box and whiskers plots of the number of MPs items per liter. MPs were detected in all the samplings at the four WWTPs, however, regarding the DBW, MPs were only found in the third sampling, which suggests a high variability over time and a greater presence of this contaminant in RWW. From the entire study, MPs appear in 22.2% of the BWDP samples, and in 77.8–88.9% of the WWTPs samples. DBW showed the lowest concentration (average of 2.0 ± 2.0 items·L^{-1}, while RWW-3 showed the highest concentration (an average of 40.0 ± 19.0 items·L^{-1}). It should also be highlighted, that the only RWW with a filtration tertiary treatment is RWW-1 and that it showed similar content to the rest of WWTPs. Among WWTPs, RWW-3 showed a significant higher concentration in COD (Table S2 of the Supplementary Material), which suggests that concentration of contaminants such as organic matter could be linked with MPs at WWTPs input. Since reverse osmosis membranes have a very small pore size, roughly in the 5–20 A range, the presence of MPs in DBW point out to a possible contamination after desalination processes. Usually, DBW is stored in tanks before distribution being there where a slight contamination by MPs might take place.

The shapes most found were microfibers of 56–4259 μm length and 5–30 μm diameter (≥ 84.4% in all cases), which accounted for 100% in RWW-4, though fragments of sizes between 87 and 611 μm were also found in samples from RWW-1, RWW-2 and RWW-3 (between 2.2 and 4.4%) as well as films with sizes between 137 and 1142 μm in RWW-1, RWW-2 and DBW (between 8.9 and 11.1%). Apart from these shapes, some microbeads (diameters in the range 56–145 μm) were also found in RWW-2 (2.2%) and RWW-3 (8.3%). Their colors were mainly blue (40.0–55.6%), black (11.1–40.0%) and transparent (15.0–33.3%), which were present in all the samples, though white, red, grey, brown and green particles were also found in some of them. Fig. 3a shows the histogram of the largest length and color distribution of the MPs found in all the irrigation waters to better appreciate both parameters. From the figure it is clear that those are also the most abundant colors and that the ones with a length in the range 200–400 μm were the most abundant (average length was 786.9 ± 812.1 μm). Fig. 4 compiles the photographs taken at the stereomicroscope of different types of MPs in both water and soil samples, in which MPs of different shapes and colors can be appreciated.

Regarding the composition, a total of 50 MPs (16.7% of total extracted MPs) were randomly selected and analyzed by μFTIR. In the particular case of marine studies, the Guidance of Marine Litter in European Seas of the

Table 1
Results of the determination of MPs in water samples from the four wastewater treatment plants and from the brackish water desalination plant.

Irrigation water	Number of analyzed samples	Total MPs	Items·L^{-1}	Shape	Color	Length (μm)
RWW-1	9	47	10.0 ± 7.2	Fibers (86.7%) Fragments (2.2%) Films (11.1%)	Blue (40.0%) Red (8.9%) Black (20.0%) Transparent (31.1%)	99–4259
RWW-2	9	45	10.0 ± 8.6	Fibers (84.4%) Fragments (4.4%) Microbeads (2.2%) Films (8.9%)	Blue (40.0%) Red (13.3%) Black (17.8%) Transparent (26.7%) Green (2.2%)	97–3498
RWW-3	9	180	40.0 ± 19.0	Fibers (88.3%) Fragments (3.3%) Microbeads (8.3%)	Blue (40.0%) Red (13.9%) Black (14.4%) Transparent (25.6%) Grey (2.2%) Brown (0.6%) White (3.3%)	56–3990
RWW-4	9	20	4.4 ± 3.5	Fibers (100%)	Blue (40.0%) Red (5.0%) Black (40.0%) Transparent (15.0%)	162–2050
DBW	9	9	2.0 ± 2.0	Fibers (89.0%) Films (11.0%)	Blue (55.6%) Black (11.1%) Transparent (33.3%)	111–2709

DBW: Desalinated brackish water; RWW: Recycled wastewater.

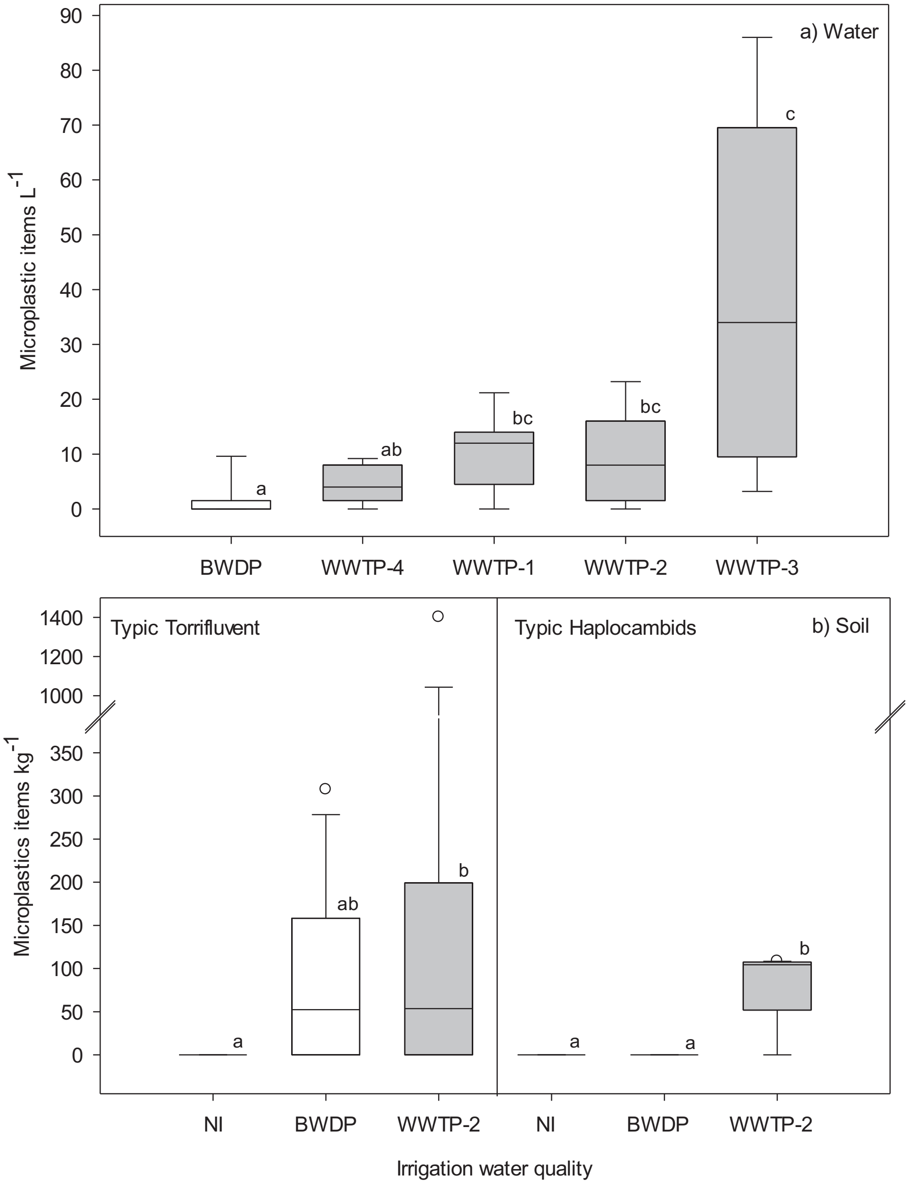

Fig. 2. Box and whiskers plot of (a) MPs concentration (items·L^{-1}) of the four RWW samples and the DBW; $n = 9$; (b) MPs concentration (items·kg^{-1}) of the soil samples under different irrigation treatments; $n = 8$. NI: No irrigation and no crop; DBW: Desalinated brackish water; RWW: Recycled wastewater. Different letters indicate significant differences ($p < 0.05$) between water qualities (ANOVA and post-hoc Tukey's test; Kuskal-Wallis test and a non-parametric Tukey-type multiple comparisons test). The boundary of the box closest to zero indicates the 25th percentile, a line within the box marks the median, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers (error bars) above and below the box indicate the 90th and 10th percentiles. Circles represent outliers.

European Commission (Galgani et al., 2013) indicates that formal identification of the polymer composition is not so critical for larger particles (> 500 μm) while a proportion of 5–10% of all samples <100 μm should be routinely checked. To the best of our knowledge, in the specific case of terrestrial environments, there does not exist such guideline; even though, we decided to consider it, despite most of the microfibers had a length higher than 500 μm. Most of the particles found (44.0%) were cellulosic, 18.0% PES, 4.0% a polypropylene (PP)-PE copolymer, 4.0% azlon, 2.0% acrylic, 2.0% nylon, 2.0% polyvinyl acetate ethylene, and 24.0% were not identified (see Fig. 5a) since the acceptable matching percentage (> 70%) could not be achieved (Galgani et al., 2013; Fernández-González et al., 2021). As previously indicated, natural fibers, like cotton, wool and linen, and semi-synthetic fibers, like rayon, viscose, and cellophane were classified as cellulosic due to the high similarity of their FTIR spectra (Cai et al., 2019; Suaria et al., 2020).

These data are the first to be reported concerning RWWs and DBW of the Canary Islands, and one of the very few already published in the whole country (Bayo et al., 2016, 2020a; Edo et al., 2020; Franco et al., 2020). Results of a revision of published literature showing MPs studies in RWWs potentially used for agricultural irrigation, are shown in Table 2. MPs shape, size and color in the present study generally agree with those previously reported and also with those published in review articles devoted to the general study of wastewater (Cheng et al., 2021; Hamidian et al., 2021), being fibers the most abundant shapes and blue one of the predominant colors. Regarding the concentration of MPs, in this study average concentrations ranged from 4.4 to 40.0 items·L^{-1}, while in the literature about RWWs potentially used for agricultural irrigation they ranged from 0.31 to 59.3 items·L^{-1} (Table 2). These differences could be related to different removal rates efficiencies accomplished by contrasting water treatment processes, but also to unlike daily water volumes treated, and load of MPs in the WWTPs inputs.

Regarding a comparison between the composition of the MPs found in this work with those previously reported in the literature, it should be first highlighted that any comparison in this sense should be taken

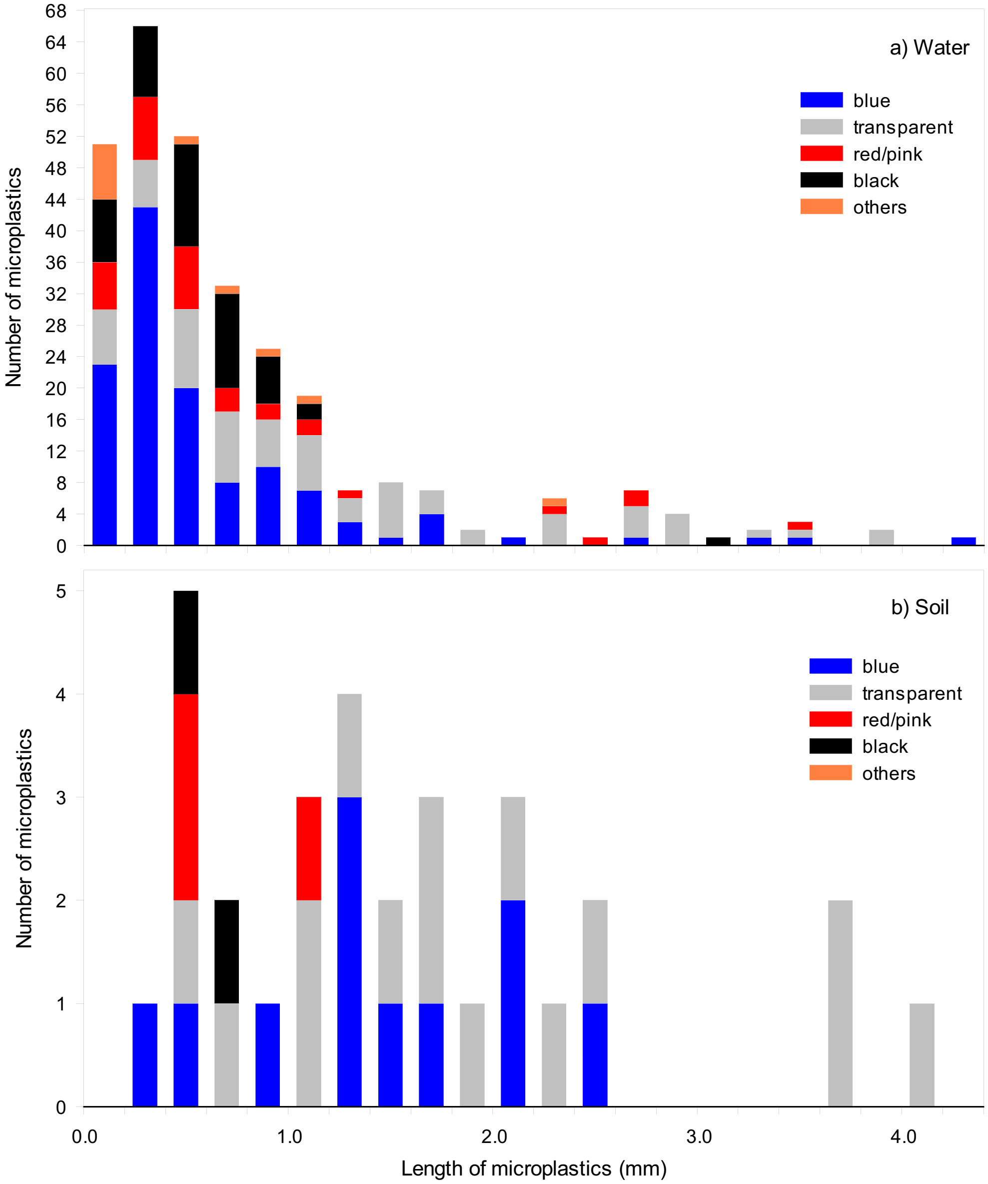

Fig. 3. Histogram of the length (largest dimension) and color distribution of the MPs found in the irrigation waters (a), and the soil samples (b).

carefully, because studies have adopted different sampling and sample treatment criteria. The materials predominantly reported in the literature have been PET, PP, PE and polystyrene (PS) which are the compositions most frequently found when films or fragments are the main shapes analyzed (Cheng et al., 2021; Hamidian et al., 2021), except for PET, which has been the main composition of microfibres (Gaylarde et al., 2021). The fact that PES accounts for 18.0% of the identified microfibers agrees with this finding. The presence of the rest of the polymers also concurs with the reported data pointed aout wastewaters are really complex and may contain plastics from different inputs. However, important amounts of cellulosic microfibers were also found (44.0%), which have not been so widely reported in the literature (in many cases the composition of the microfibers are not fully explored, or cellulosic fibers are not considered), except in some specific cases in which, for example, rayon has been found at high percentages (Cheng et al., 2021; Hamidian et al., 2021). In our study, if it can be assumed that the 16.7% of MPs composition analyzed represent the total amount of MPs extracted, subtracting the amount of cellulosic fibers (44.0%) from the total, the concentrations of MPs would get much closer to those reported in the literature (Table 2). Since it is clear that these fibers have an anthropogenic origin (natural fibers are also frequently dyed and released with wastewaters), this particular issue should be further investigated though, in the specific case of the Canary Islands, that have a high number of wastewater discharge points, important amounts of cellulosic fibers have been found in the marine environment, probably as a results of such discharges (Villanova-Solano et al., 2022; Sevillano-González et al., 2021).

3.3. Microplastic occurrence in soil samples

Fig. 2b and Table 3, compiles the content and classification of the MPs found it the two types of soils assessed under irrigation with RWW-2 and DBW, while Fig. 3b shows the distribution of colors and lengths of the MPs extracted from soils. As can be seen, MPs were not found in any of the non-irrigated/non-cultivated soils, while those irrigated with RWW showed the highest content, ~ 238 and 79 items·kg^{-1} for soils TT and TH, respectively. In the TT soil irrigated with DBW the average concentration was ~91 items·kg^{-1}, whereas in the analogous TH one, no MPs were extracted. From the total soil samples analyzed 0, 25, and 62.5% of samples non-irrigated/non-cultivated, irrigated with DBW, and irrigated with RWW presented MPs, respectively. These data could suggest that there are no

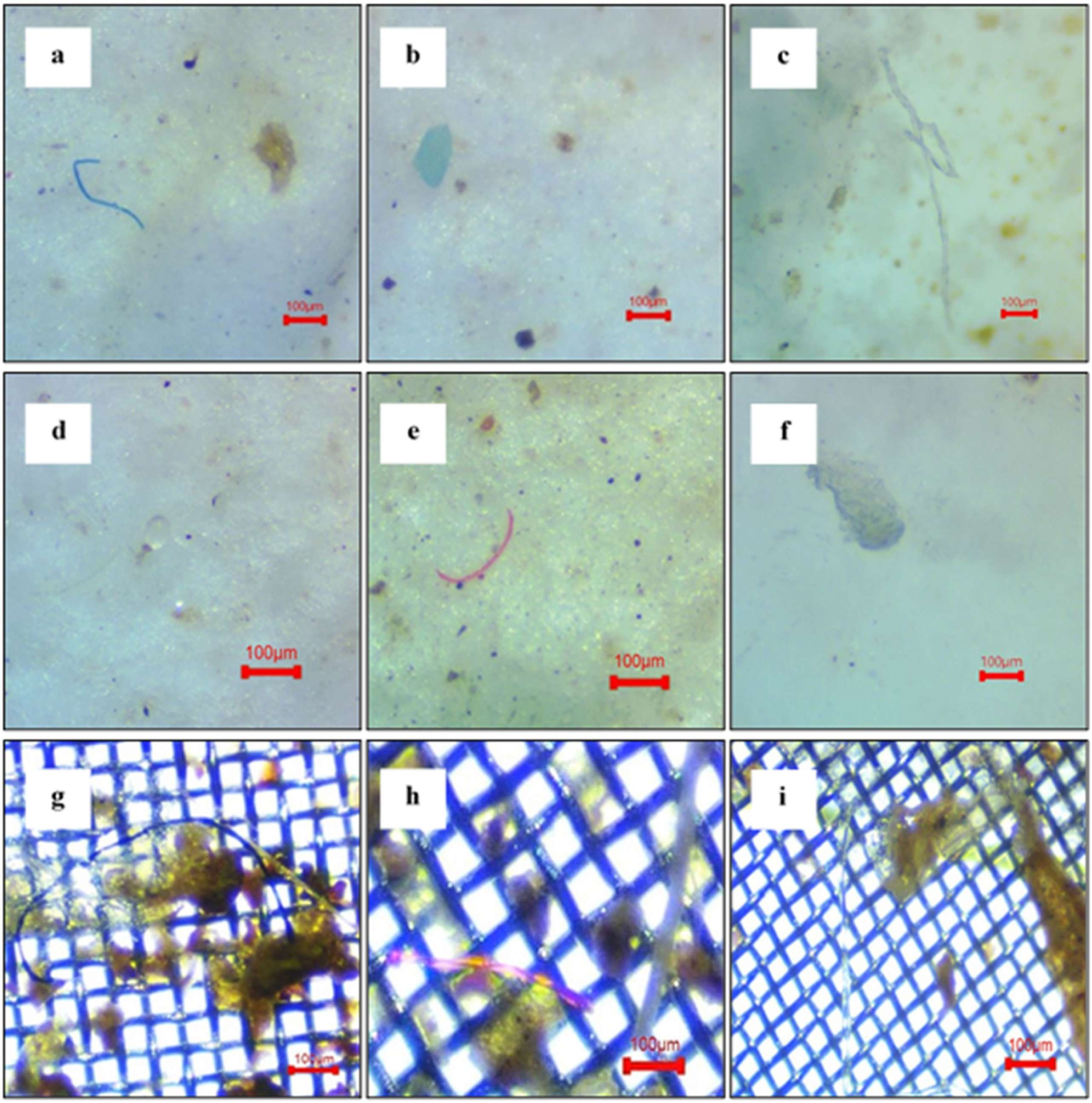

Fig. 4. Images of MPs detected in irrigation waters and soil samples: (a) Blue fiber from RWW-3 in sampling date 07/14/2021, (b) Blue fragment from RWW-3 in sampling date 07/14/2021, (c) Transparent fiber from RWW-1 in sampling date 05/21/2021, (d) Transparent microbead from RWW-2 in sampling date 07/14/2021, (e) Red fiber from RWW-3 in sampling date 06/29/2021, (f) Transparent film from RWW-2 in sampling date 05/21/2021, (g) Blue fiber from TT soil irrigated with RWW-2 in sampling date 02/17/2021, (h) Red fiber from TT soil irrigated with RWW-2 in sampling date 02/17/2021 and (i) Transparent fiber from TH soil irrigated with RWW-2 in sampling date 02/17/2021. RWW: Recycled wastewater; TT: Typic Torrifluvents; TH: Typic Haplocambids.

other significant MPs sources, apart from agricultural activities, in these areas not subjected to a high anthropic pressure. Considering that soils with RWW and DBW irrigation have been managed in an identical way (e.g., same crop, fertilization, irrigation doses), observed differences in MPs contamination could be attributed to a higher MPs load in water from WWTPs in comparison with water from BWDP (Fig. 2a). Despite differences in soil TT and TH physical characteristics (e.g., soil TT has a coarse texture and more macroporosity allowing a rapid water infiltration, whilst soil TH has a fine texture, high microporosity, and usually surface sealing, decreasing the infiltration rate), no significant differences were found between both soils in MPs concentration when irrigated with RWWs (Fig. 2a).

The amount of MPs reported in this study (~ 79–238 items·kg^{-1}) is generally lower than published data in agricultural soils with several MPs sources, for example Zhang and Liu (2018) ~ 7100–42,960 items·kg^{-1}; Corradini et al. (2019) ~ 600–10,400 items·kg^{-1}; Zhou et al. (2019) ~ 96,000–690,000 items·kg^{-1}; Chen et al. (2020) ~ 320–12,560 items·kg^{-1}; van den Berg et al. (2020) ~ 930–3060 items·kg^{-1}; and Zhou et al. (2020a) ~ 263–571 items·kg^{-1}.

Table S4 of the Supplementary Material compiles data of articles published in the literature during the last two years (2020-2021) related with MPs pollution in agricultural soils. As can be seen in that table, MPs items per kg of soil ranged from 3.7 to 40,800, with reported MPs concentrations similar or even higher than those found in this study, but usually focused

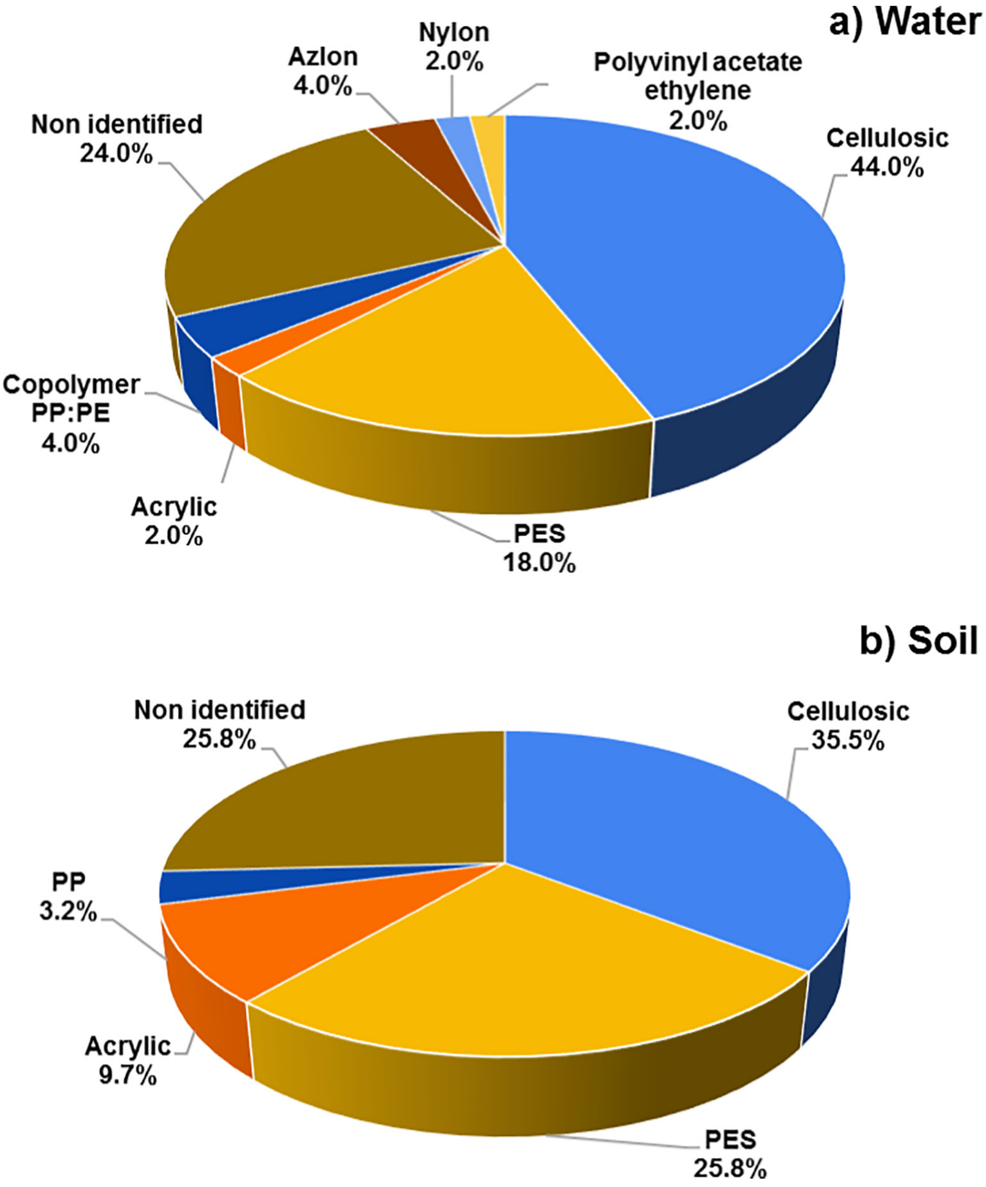

Fig. 5. Distribution of the composition of MPs found in the irrigation RWWs (a) as well as in the irrigated soil samples (b). RWW: Recycled wastewater; PP: Polypropylene; PE: Polyethylene; PES: Polyester.

just on film and fragments shapes determination but not on fibers. Besides, such data should be compared with care, since the same extraction methods have not been applied in all of them. Even though, almost no papers have assessed MPs pollution in soils where RWW were the main or unique potential MPs source; therefore, effects of different practices (e.g., compost or sewage sludge application, mulching films, greenhouses, irrigation with different water qualities) can overlap. As previously mentioned, low intensity agricultural conditions in our study made potential MPs source less abundant and then, lower contamination can be expected. Attending to the MPs load in RWW-2 (∼ 10 items·L^{-1}) and the volume of water applied for crop production in soils of this study (approximately 2500 mm·year^{-1}) the soil MPs concentration should be much higher than that found (∼ 4400 items·m^{-2} and 14,300 items·m^{-2}, in a 5 cm thick layer, for soil TH and TT respectively). Such fact could be linked to weathering processes, drag with water into deeper soil layers, transport and alteration by soil fauna, and/or re-suspension and transport for wind and runoff, but also to MPs retention processes during water distribution from the WWTPs, and particularly by different component of the irrigation systems in the field (e.g., cleaning filters, drippers). Therefore, sampling water immediately after passing irrigation system could be a better approach in order to evaluate the real amount of MPs reaching the soils.

As can be seen in Table 3 and Fig. 3, all MPs found in soils were fibers with lengths in the range 310–4069 μm, mainly blue and transparent mostly matching with the characteristics of MPs in irrigation waters (Table 1). Fig. 4 shows several photographs of some of the MPs found in the soil samples.

Concerning the composition (see Fig. 5b), 100% of the particles founds in soils were analyzed by μFTIR as a result of the relatively low number of microfibers present ($n = 31$). Analysis revealed that 35.5% were cellulosic, 25.8% PES, 9.7% acrylic, 3.2% PP and 25.8% could not be identified since the acceptable matching percentage (> 70%) could not be achieved (Galgani et al., 2013; Fernández-González et al., 2021). These results also agree with composition of MPs in the irrigation water where cellulosic and PES are the materials more frequents found, reinforcing the hypothesis that irrigation waters, particularly RWW, could represent a source of MPs contamination in soils.

Table 2

Previous studies in the literature about MPs determination in the final wastewater treatment plants effluent used for irrigation purposes and comparison with this study.

Country	Treatment	Capacity (m^3 d^{-1})	Analytical method	Items·L^{-1}	Dominant shape	Dominant size	Dominant color	Dominant polymers	Reference
Spain	Secondary (activated sludge)	$3.5 \cdot 10^4$	Filtration	0.31	Fragments (60.5%)	600–800 μm	–	LDPE, PTFE, Acrylic, PES, PET, PTFE	Bayo et al. (2020b)
Spain	Secondary (MBR)	$1.2 \cdot 10^4$	Filtration	0.92	Fibers (90.8%)	1–2 mm	–	MUF	Bayo et al. (2020a)
	Tertiary (RSF)	$1.8 \cdot 10^4$	Filtration	1.08	Fibers (96.8%)	1–2 mm	–	LDPE, NYL, PV	
Canada	Secondary	$8 \cdot 10^7$	Fenton digestion; filtration	1.76	Fibers (81%)	–	–	PE, PET, PMMA	Prajapati et al. (2021)
Israel	Tertiary (RSF)	$3 \cdot 10^4$	Fenton digestion; NaCl flotation; filtration; staining with Rose-Bengal solution	1.97–7.30	Fibers (91%)	1000–2000 μm	Black (50%) Blue (20%)	PE, PET, PVC, PP	Ben-David et al. (2021)
Spain	Tertiary (RSF)	$6.5 \cdot 10^3$	Filtration	1.38	Fibers (92.9%)	1–2 mm	Grey (48.9%) Beige (19.2%)	LDPE, HDPE, Acrylic	Bayo et al. (2021)
Iran	Secondary	(−)	H_2O_2 digestion; NaI flotation; filtration; staining with Rose-Bengal solution	0.42	Fibers (78%)	37–300 μm	–	PES, Acrylic, NYL, PE	Petroody and Hashemi (2021)
Mauritius	Tertiary	$5.5 \cdot 10^4$–$5.9 \cdot 10^4$	H_2O_2 digestion; filtration	59.3	Fibers (54%)	0.50–0.25 mm	Brown (41%)	PA, PE, EVA	Ragoobur et al. (2021)
Spain	Secondary (activated sludge)	$6 \cdot 10^1$ – $4 \cdot 10^3$	Filtration	4.4–40	Fibers (≥ 84.4%)	200–400 μm	Blue (40%) Transparent (25%)	PES, PP:PE, Acrylic	This study
	Tertiary (RSF)	$4.2 \cdot 10^3$	Filtration	10.0	Fibers (86.7%)	200–400 μm	Blue (40.0%) Transparent (31.1%)	PES, Azlon, NYL	

RFS: Rapid sand filtration; MBR: Membrane bioreactor technology; LDPE: Low density polyethylene; HDPE: High density polyethylene; PE: Polyethylene; PP: Polypropylene; MUF: Melamine; PTFE: Teflon; PET: Polyethylene terephthalate; PES: Polyester; NYL: Nylon; PES: Polystyrene; PVC: Polyvinyl chloride; PMMA: Poly methyl methacrylate; PV: Polyvinyl; PP:PE: Polypropylene and polyethylene copolymer.

4. Conclusions

The determination of MPs in RWW and DBW from Fuerteventura island revealed the presence of cellulosic and PES blue and transparent fibers, a pattern that was maintained in the two types of soils (sandy-loam and clay-loamy) irrigated with those water qualities during four years. However, since the DBW contained significantly less amounts of MPs, DBW irrigated soils also showed a less concentration compared to RWW irrigated ones. No MPs were found in soils not irrigated and not cropped during the same period. This issue, together with the fact that practices such as mulching film, fertilization with sewage sludge or compost were not applied to soils, allow to establish a clear relationship between MPs presence in RWW and irrigated soils.

In the specific case of arid territories with limited water supply as Fuerteventura Island, where agricultural production significantly relies on the use of final effluents from numerous different size and technologies WWTPs, it is desirable to develop a long-term study for assessing their MPs removal efficiency in order to analyze possible actions to decrease the MPs load reaching the agricultural soils and impacting soil quality and productivity. Notably, the presence of MPs in DBW obtained by reverse osmosis techniques that use membrane with very fine pore size (5–20 A), suggest a MPs contamination post desalination processes during water storage or distribution that should be assessed.

This work provided the first data concerning MPs presence in RWW and DBW as well as in soils from the Canary Islands (Spain) and one of the very few articles in the literature in which a clear relation between MPs contained in irrigation waters and soils could be established.

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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Table 3

Results of the determination of MPs in the Typic Torrifluvents and Typic Haplocambids soils under different irrigation treatments.

Soil TT (sandy-loam)	Analyzed samples	Total MPs found	Items·kg^{-1}	Shape	Color	Length μm	Polymer types %
Irrigated with RWW-2	8	18	238.3 ± 478.1	Fibers (100%)	Blue (33.3%) Transparent (38.9%) Red (16.7%) Black (11.1%)	310–4069	Cellulosic (38.9%) PES (27.8%) Acrylic (11.1%) PP (5.6%) Unidentified (16.7%)
Irrigated with water from DBW	8	7	91.1 ± 116.3	Fibers (100%)	Blue (57.1%) Transparent (42.9%)	526–3701	PES (28.6%) Acrylic (14.3%) Unidentified (57.1%)
Non-irrigated/non-cultivated area	8	0	0.0 ± 0.0	–	–	–	–
Soil TH (clay-loamy)							
Irrigated with water from RWW-2	8	6	79.5 ± 49.1	Fibers (100%)	Blue (16.7%) Transparent (83.3%)	1009–3668	Cellulosic (66.7%) PES (16.7%) Unidentified (16.7%)
Irrigated with water from DBW	8	0	0.0 ± 0.0	–	–	–	–
Non-irrigated/non-cultivated area	8	0	0.0 ± 0.0	–	–	–	–

TT: Typic Torrifluvents; TH: Typic Haplocambids; DBW: Desalinated brackish water; RWW: Recycled wastewater.

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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.2021.152830.

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