

Article

Effect of Phytoremediated Port Sediment as an Agricultural Medium for Pomegranate Cultivation: Mobility of Contaminants in the Plant

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Abstract: Although the dredging of ports is a necessary management activity, it generates immense quantities of sediments, that are defined by the European Union as residues. On the other hand, the relevant peat demand for plant cultivation compromises its availability worldwide. In this context, the present work wanted to find an alternative substrate in order to replace and/or reduce the use of peat in agriculture, through the study of the suitability, concerning the exchange of substrate–plant–water pollutants, of the dredged remediated sediments as a fruit-growing media. Forty-five pomegranate trees (*Punica granatum* L. cv “Purple Queen”) were cultivated in three types of substrates (100% peat as a control, 100% dredged remediated sediments and 50% both mixed). The metal ion content and pesticide residues were analysed in the different plant parts (root, stem, leaves and fruits) and in drainage water. The results showed a limited transfer of pollutants. All the pollutants were below the legal limits, confirming that the dredged sediments could be used as a growing media, alone or mixed with other substrates. Thus, the results point out the need to open a European debate on the reuse and reconsideration of this residue from a circular economy point of view.

Keywords: fruit production; heavy metal; pesticide residue; *Punica granatum* L.; port sediment



Citation: Martínez-Nicolás, J.J.; Legua, P.; Hernández, F.; Martínez-Font, R.; Giordani, E.; Melgarejo, P. Effect of Phytoremediated Port Sediment as an Agricultural Medium for Pomegranate Cultivation: Mobility of Contaminants in the Plant. *Sustainability* **2021**, *13*, 9661. <https://doi.org/10.3390/su13179661>

Academic Editor: Emanuele Radicetti

Received: 3 August 2021

Accepted: 24 August 2021

Published: 27 August 2021

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1. Introduction

Each year, important quantities of sediments are dredged in maritime ports. The dredged activity can be defined as the operations set needed for the extraction, transport and dumping of underwater materials, either in the sea, rivers or lakes. Although necessary to guarantee the cargo boat transit, these operations directly or indirectly have negative effects for the environment [1].

The dredged sediments usually present a very heterogeneous composition, with different physical, chemical and biological characteristics and properties depending on the specific port anthropogenic activities [2]. The most common pollutants identified on dredged port sediments are heavy metals [3] and polycyclic aromatic hydrocarbons (PAHs) [4], and due to this, the European legislation defines them as contaminated residues, thereby restricting their use [5].

However, recent studies indicated that the vast majority of the amounts of sediment, around 90%, are not contaminated [6]. In these cases, the dredged sediment could be considered a resource rather than a residue able to reuse and revalorise from the circular economy perspective [7]. Thus, recent multidisciplinary scientific studies have reported, demonstrated and confirmed the viability of obtaining a new safe substrate from the treated dredged sediments related to the amounts of contaminants present in it [6,8–10].

On the other hand, during the last five decades, the peat has been the most-utilised substrate for nursery activity due to its optimal physicochemical characteristics for many plant species [11–13]. However, nowadays, the agricultural use of peat is threatened due to its overexploitation, which increases global agricultural conflicts. Therefore, the high agricultural production demands for the populations with the peatlands on the brink of collapse motivate to find new substrates that allow to decrease, or even replace, the peat used in agriculture [11]. In this context, many advancements on the residue and/or by-products' reuse and revalorisation for agriculture use have been observed. Different materials have been applied successfully as peat substitutes for crop production, such as rice husks, almond shells and coconut fibres, among others, and composted materials (animal manure and agro-industrial residues) and/or alternative materials as biochar [14–22].

The general objective of this investigation was to verify the dredged remediated sediment's suitability, alone and/or mixed with peat, to be an alternative culture media to the traditional substrate. The sediment's suitability as an agricultural substrate was studied from the point of view of the mobility/transfer of pollutants and minerals between the substrate–plant–water system, in order to identify possible environmental and consumption risks. The experimental assays were carried out in pomegranate trees (*Punica granatum* L. cv “Purple Queen”) due to both their commercial relevance and to the great technical-scientific knowledge presented by the responsible research group.

2. Materials and Methods

2.1. Properties of the Dredged Remediated Sediment

The dredged remediated sediment (DRS) used in this study came from Livorno Port (Italy) and was previously phytoremediated for three years in an already published study [4,23,24]. At the end of the phytoremediation process, the DRS presented a good nutrient content, good biological activity and a low contamination level to be used as culture media according to the Spanish legislation on growing substrates for agricultural uses (Royal Decree 865/2010 of 2 July [9,25]). The initial DRS characterisation used in this study is shown in Table S1 of the Supplementary Materials.

2.2. Plant Material and Experimental Design

This experiment was carried out over two years (2016 and 2017) in an experimental crop plot at the Higher Polytechnic School of Orihuela (Miguel Hernandez University), located in the Southeast of Spain (38°04' N, 0°58' W, 26 m above sea level). The average annual temperature was 19 °C, with moderate winters (minimum of 11 °C in January) and warm summers (maximum of 28 °C in August). The mean annual precipitation, during the experimental time (2 years), was 300 mm, and most of this precipitation was recorded in the spring and autumn seasons. The area presented high insolation, with approximately 3000 h of sunshine per year. The highest sunshine percentage was registered during the summer months (more than 352 h in July) and the lowest in December and January.

The plant material used was pomegranate tree (*Punica granatum* L., cv “Purple Queen”) acquired from a commercial nursery. The plants were provided in 1.5 L polyethylene bags, with coconut fibre as a substrate. The plants were ca. 70 cm high and the trunk's diameter was ca. 0.6 cm. After transplanting, which was conducted on 5 May 2016, the pomegranates were shortened to 40 cm in height. The pomegranate trees were transplanted to 40 L polyethylene pots with three different substrates: (i) 100% peat (Pt) as a substrate control, (ii) 100% dredged remediated sediments (DRS) and (iii) 50% mixture (Pt-DRS). The main physicochemical characteristics of the commercial peat used as the control substrate were: pH 5.87, bulk density 0.58 g mL⁻¹, organic material: 10% ashes, cation exchange capacity (CEC) 120 m_{eq} 100g⁻¹, total N 0.13%, P₂O₅ 150 mg L⁻¹, K₂O 220 mg L⁻¹, MgO 144 mg L⁻¹.

For each substrate, three replicates comprised of five trees each were used, which were distributed using a completely randomised block design (15 trees per substrate). A total of 45 pomegranate trees were employed. In order to avoid a border effect, 40 trees grown

in pots with peat were placed surrounding the treated trees (Figure S1, Supplementary Materials). The crop management was homogeneous for the three substrates, aiming to verify and study the impact of the DRS as a growth media.

2.3. Hydric Monitoring and Drainage Characterisation

The irrigation water was regulated for the three substrates studied (Pt, Pt-DRS, DRS) by a high-frequency irrigation system with drippers set at 2 L h⁻¹ per plant. The pot soil moisture was monitored daily by Watermark[®] soil moisture sensors (200SS-Irrrometer, Riverside USA) installed at the root level (10 cm from the dripper and 15 cm of depth). For each substrate, three soil moisture sensors were employed to identify possible variations. The irrigation programming was defined based on the results obtained by the irrometer sensors.

During the assay, the pH and electrical conductivity (EC) of the irrigation water were monitored and maintained constant (2.2 dS m⁻¹), aiming to identify the potential variations in drainage water samples due to the interaction with the substrates. The drainage water was collected separately for each experimental block, and pH and EC were periodically monitored by a multi-parameter analyser (Consort C860, Topac Inc., Cohasset, MA, USA). The drainage volume was monitored for each substrate through a graduated measuring cylinder (2000 mL maximal capacity, polyethylene, non-sterile). Due to the sediment characteristics, pesticide residues, metal ion and macro- and micro-elements were analysed only in the DRS drainage, since DRS drainage predictably will show the highest values when compared with the other substrates and, therefore, is potentially dangerous. Therefore, for macro- and micro-elements, metal ions and pesticide residues' determinations, 30 mL of drainage water samples were collected in a 50 mL maximal volume polyethylene container. The drainage samples were conserved at constant temperature (−5 °C) until their use for chemical analysis, as explained below. The pesticide residue was determined following the UNE-EN 15662:2019, EPA 1668C 2010 and EPA 8015C 2007 methods [26–28]. The pesticide analyses were carried out in triplicate by an accredited laboratory (KUDAM S.L.U, Spain. www.kudam.com (accessed on 20 August 2021)).

2.4. Plant Samples and Pollutant Determination

The pomegranate fruits were collected manually once fruits reached the maturity status. Five pomegranate fruits per tree for each substrate studied were used. The fruit samples were collected from all tree orientations. For the fruit samples, seeds and peel were manually separated the same day of the collection and freeze-dried (Christ Alpha 2-4 LSCbasic, Martin Christ, Germany) until analysis.

In addition, at the end of the growing stage, pomegranate trees were sacrificed and divided into plant parts: root, stem and leaves. The pomegranate plant parts' samples were oven-dried (ED23, Binder, Germany) at 60 °C until constant weight, and stored in a glass desiccator with silica gel (LbG, Labbox, Spain) until analysis. All the vegetal samples were analysed to quantify the mineral elements, metal ions and the presence of pesticide residues, as described below.

2.4.1. Mineral Ions' Determination

For the anions' analysis (nitrate, sulphate, nitrate, bromine and fluorine), the ion chromatography (IC-Plus) method with a conductivity detector was used (883 Basic IC plus-Metrohm, Switzerland), following the methodology described by Martínez-Nicolás et al. [9].

Carbonates and bicarbonates were determined through a sample exchanger for measuring the pH and conductivity (814 USB Sample Processor, METROHM), and the data were analysed by the software Tiamo 2.3. METROHM. The spectrometer UV-Visible VAR-IAN CARY 50 was used to quantify the N from ammonia according to the 4500-NH3 Standard Methods methodology [29].

2.4.2. Metal Ions' Content

The determination of metal ions (manganese (Mn), iron (Fe), zinc (Zn), copper (Cu), molybdenum (Mo), lead (Pb), cadmium (Cd), nickel (Ni), chromium (Cr), mercury (Hg), cobalt (Co), antimony (Sb), arsenic (As), selenium (Se), aluminium (Al), beryllium (Be), tin (Sn), strontium (Sr), silver (Ag), thallium (Tl) and vanadium (V)) was conducted by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7700-E and Agilent 7700-X). The samples' preparations were performed according to the 200.8 EPA and UNE-E 13804:2013 methods [30,31], and the analyses were carried out by a certified laboratory (KUDAM S.L.U, Spain. www.kudam.com (accessed on 20 August 2021)).

2.4.3. Pesticide Residues in Plant Material

According to the chemical specification of the pesticides, two different techniques were employed to determine and quantify pesticides in pomegranate plant parts. Thus, while a general pesticide identification was determined following the UNE-EN 15662:2019 method [26] by liquid chromatography coupled with quadrupole mass spectrometry (LC-MS/QqQ, Bruker Evoq Elite), the determination of polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCS) and specific fumigant residues was carried out by gas chromatography coupled with a triple quadrupole mass spectrometer (GC-MS/QqQ, Bruker Scion TQ), according to the EPA 1668C 2010 and EPA 8015C 2007 methods [27,28]. For both methodologies, the pollutant identification was performed by a retention time comparison with standard calibration solutions (>99.7%, Sigma Aldrich). The pesticide analyses were carried out in triplicate by an accredited laboratory (KUDAM S.L.U, Spain. www.kudam.com (accessed on 20 August 2021)).

2.5. Statistical Analysis

Statistical analyses were performed using SPSS 24.0 for Windows (SPSS Science, Chicago, IL, USA). A basic descriptive statistical analysis was followed by an analysis of variance (ANOVA) for mean comparisons. The method used to discriminate among the means (multiple range test) was Tukey's HSD test ($p \leq 0.05$).

In addition, a principal component analysis (PCA) was also performed for the three substrates analysed, starting with the analysis of metal and metalloid ions obtained from the inedible pomegranate parts (root, stem and leaves), followed by the pomegranate fruit, separating the edible part (seed) from the peel and the carpellary membranes of the fruit.

3. Results

3.1. Irrigation and Drainage Monitoring

The results of monitoring irrigation and plant drainage volume are shown in Figure 1. Even though the plants were in a homogeneous climatic condition since May (start of experiment date), differences in the water needed for each substrate were observed during the second experimental year. July, August and September of 2017 were the months with the highest water needs and with significant differences between the substrates (Figure 1B). Note that, in the Mediterranean climate, the summer is between June and September, with high temperatures and a dry environment, so during these months, due to the plant evapotranspiration increase, it was necessary to apply more irrigation water ($P_t > P_t - DRS > DRS$). Small differences in irrigation needs were also observed at the beginning of the summer of the second year, highlighting the different behaviour of the substrates. In general, no significant differences were observed for the percentage of drainage water obtained for each substrate and month. In 2016 (first year of treatment), the volume of water drained was more irregular than in 2017. With the exception of September and October 2016, where the highest drainage volumes (40%) were observed (Figure 1C), during the rest of the trial, the percentage of drainage was more constant, at around 20%.

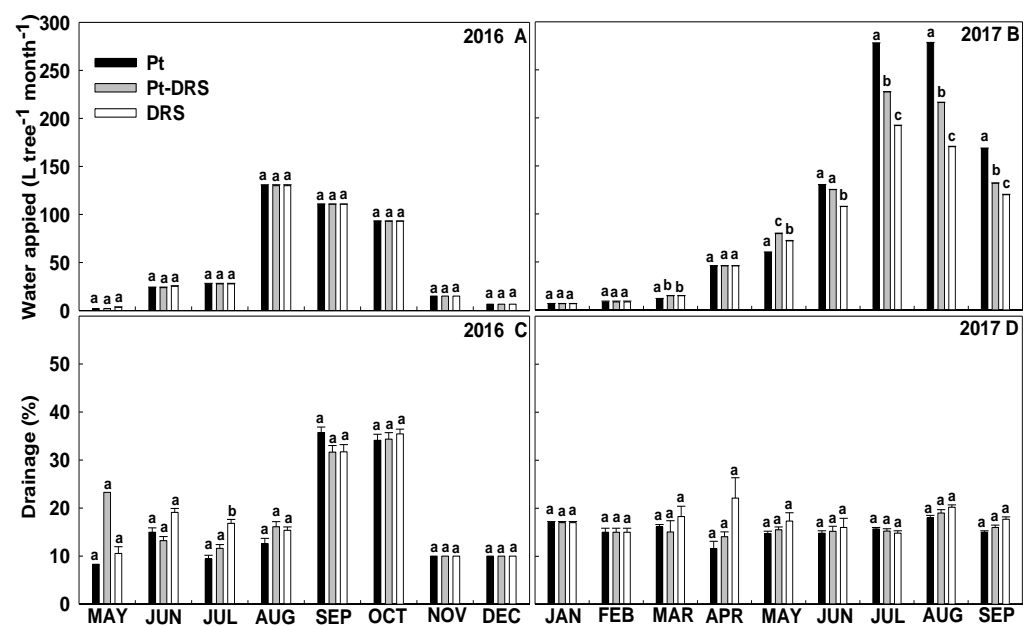


Figure 1. Monthly average water applied ($\text{L tree}^{-1} \text{ month}^{-1}$) in 2016 (**A**) and 2017 (**B**), and plant pot drainage (%) in 2016 (**C**) and 2017 (**D**). Each bar corresponds to the average of the trees of each treatment in the month. Different letters on top of bars indicate significant differences according to Tukey's HSD test ($\rho \leq 0.05$).

3.2. Plant Pot Drainage Characterisation

Aiming to identify and study the ionic exchange between the substrate–plant–water system, as well as its impact on plant development, the drainage water was physicochemically characterised throughout the trial. The electrical conductivity (EC) of plants' drainage water was variable during the first year of cultivation, with values around $>4 \text{ dS m}^{-1}$, where June 2016 presented the highest values (6 dS m^{-1}). During the second year, EC values' stability was observed, at around 3.5 dS m^{-1} . The greatest variability was detected for July 2017, with significant differences between the substrates. In general, EC values were higher during 2016 than 2017 (Figure 2A,B). The same trend of stabilisation during the second experimental year was observed for the pH values in the drainage of the different substrates. Therefore, during the first experimental year, the Pt drainage presented lower pH values (pH = 6) than the other two substrates, with pH values around 8 (Pt < DRS < Pt-DRS) until September. From September until the end of the experiment, the drainage pH values were stabilised and equalised for all the substrates (pH = 8), without significant differences (Figure 2C,D).

Since peat is a widely known and studied substrate, the characterisation and quantification of both micro- and macro-elements and metal ions were only carried out in the drains of plants grown with DRS, aiming to identify the variations on the dredged sediment composition, its real impact on the plants and also the exchanges and ionic transfers within the plant–substrate–water system. As expected, mainly due to the washing out effect by plant irrigation action, the values of macro- and micro-elements were higher in 2016 than in 2017, with the exception of carbonates that remained constant and ammoniacal nitrogen that slightly increased (Table 1).

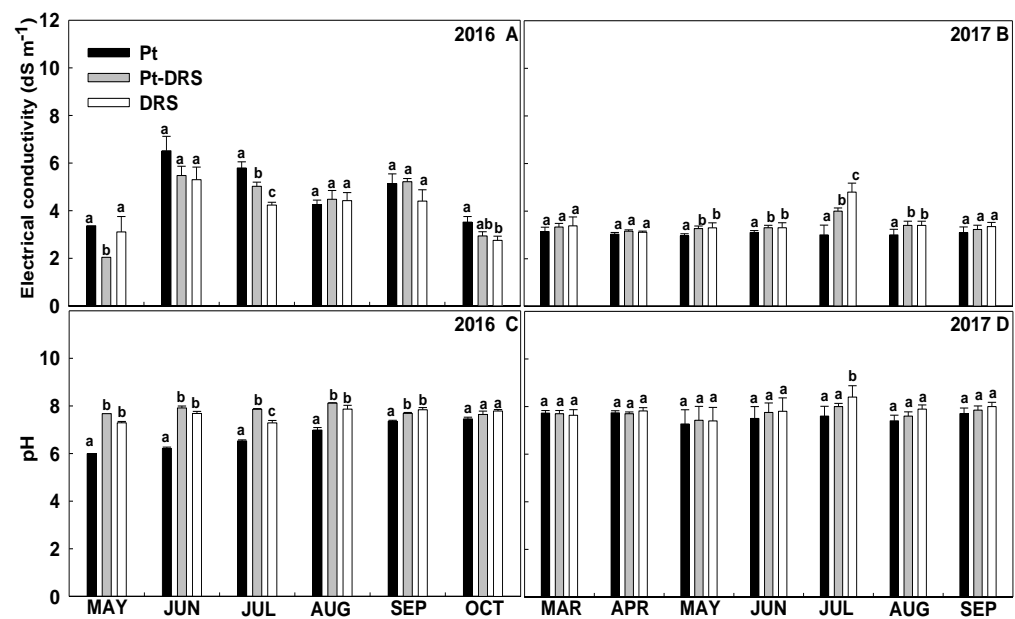


Figure 2. Monthly average electrical conductivity (dS m^{-1}) in 2016 (A) and 2017 (B), and pH in 2016 (C) and 2017 (D). Each bar corresponds to the average of the trees of each treatment in the month. Different letters on top of bars indicate significant differences according to Tukey's HSD test ($\rho \leq 0.05$).

Table 1. Macro- and micro-elements' characterisation presented in the drainage water of the pomegranate plants grown on the DRS substrate during the two years of the experimental trial.

Element	(mg L^{-1})	
	2016	2017
Sodium (Na^+)	197	119
Potassium (K^+)	412	266
Calcium (Ca^{2+})	286	152
Magnesium (Mg^{2+})	78.8	44.5
Boron (B^{3+})	1.53	0.784
Chlorides (Cl^-)	304	215
Sulphates (SO_4^{2-})	418	400
Carbonates (CO_3^{2-})	<5.00	<5.00
Bicarbonates (HCO_3^-)	268	168
Nitrates (NO_3^-)	909	269
Ammoniacal nitrogen (NH_4^+)	<0.100	0.331
Phosphates (H_2PO_4^-)	39.2	28.4
Element	($\mu\text{g L}^{-1}$)	
	2016	2017
Manganese (Mn)	197	119
Iron (Fe)	412	266
Zinc (Zn)	670	660
Copper (Cu)	183	131
Molybdenum (Mo)	1.53	0.784

The metal ions' concentration in the drained water of DRS plants were all well below the Maximum Permissible Level (MPL) established by specific Spanish legislation, as shown in Table 2. In addition, the sum of the fractions analysed with respect to the MPL values was much lower than the thresholds allowed by the current legislation. Thus, in the first year, it was only 4.2% ($3 \mu\text{g L}^{-1}$), decreasing until 3.1% in the second year (Table 2). These

values complied with the Public Water Resources Regulation, approved by the Spanish Royal Decree 849/1986, as well as the European legislation 2008/105/CE.

Table 2. Metal ions' content in the drainage water of the pomegranate plants grown on the DRS substrate during the two years of the experimental trial and their comparison with the sum of the fractions establish by the legal Maximum Permissible Level (MPL).

Metal Ion	$(\mu\text{g L}^{-1})$				
	2016	2017	MPL	Fraction 2016	Fraction 2017
Cadmium (Cd)	17.2	11	500	0.0344	0.0220
Chromium (Cr)	<10.0	<10.0	500	0.0200	0.0200
Nickel (Ni)	41	16.5	10,000	0.0041	0.00165
Mercury (Hg)	<0.20	1.83	100	0.0020	0.0183
Lead (Pb)	7.71	2.58	500	0.01542	0.00516
Copper (Cu)	183	131	10,000	0.0183	0.0131
Zinc (Zn)	670	660	20,000	0.0330	0.0335
			Σ total	0.12722	0.09371
			MPL	3	3

Note: MPL is Maximum Permissible Level established by the Spanish Regulation of the Hydraulic Public Domain (Royal Decree 849/1986).

A summary of the identified pollutants is shown in Table 3. Of the 623 pesticide residues analysed in the DRS plants' drainage water, only 6 were detected during the 2 years of experimentation. In 2016, Acetochlor, Anthraquinone and Simazine were detected, while in 2017, only Acenaphthylene, Phenanthrene and Fluorene were identified. In all cases, the pesticide residues were below the MPL. Thus, in 2016, the sum of all the pesticide residues detected was 20.7% of the MPL (0.05 mg L^{-1}), while in 2017, the values decreased to 0.21% (Table 3), according to the Public Water Resources Regulation (Spanish Royal Decree 849/1986) and European legislation (2008/105/CE).

Table 3. Pesticide residues detected in the water drainage of plants grown with DRS.

Pesticide Residues	$(\mu\text{g L}^{-1})$	
	2016	2017
Acetochlor	10	ND
Anthraquinone	0.13	ND
Simazine	0.23	ND
Acenaphthylene	ND	0.025
Phenanthrene	ND	0.060
Fluorene	ND	0.022
Total amount	10.36	0.107
MPL	50	50

MPL: Maximum Permissible Level established by the Spanish Regulation of the Hydraulic Public Domain (Royal Decree 849/1986). ND: Not Detected.

3.3. Ions' Content and Pesticide Residues' Determinations in Non-Edible Plant Material

The ion concentrations were determined in the non-edible parts (root, stem and leaves) for the pomegranates cultivated on the tested substrates (Pt, Pt-DRS and DRS), as shown in Table 4. The analyses on the leaves were carried out during the two years of the experiment (2016 and 2017), while the analyses of the pomegranates' roots and stems were only conducted at the end of the experiment with the sacrifice of the plant. Mn, Fe, Zn, Al and Sr were the metal ions most abundant in all non-edible pomegranate parts, with concentration ranges of 20–138 ppm, 47.9–3350 ppm, 24–124 ppm, 49–4710 ppm and 35.4–139 ppm, respectively. For the different non-edible parts, iron, aluminium and strontium content were higher in root > leaf > stem, while manganese and zinc content

were root > stem > leaf and stem > root > leaf, respectively. Related to the substrate type, DRS presented the highest concentration for most of the metal ions, followed by Pt-DRS > Pt. As expected, the DRS roots presented the most elevated values of Mn (138 ppm), Fe (3350 ppm), Zn (112 ppm), Al (4710 ppm) and Sr (139 ppm), probably due to the direct contact with the substrate. Given that this plant part is not used for food, the metal ions' content should not be a problem, because almost all the metal ions detected were below the limit values, and only Cd (0.679 ppm) and Pb (6.88 ppm) concentrations were above those allowed for food (Codex alimentarius 1995; European Regulation 1881/2006). However, the metal ions' content can lead to an inhibition in the growing plant, so it must remain within plant tolerance limits. No pesticide residues were detected in the non-edible pomegranate parts during the assay.

Table 4. Metal ions' content (ppm) in the non-edible pomegranate parts (root, stem and leaf) cultivated on the different substrates (Pt, DRS and Pt-DRS) during the two experimental years (2016 and 2017). Different letters within a column indicate significant differences by Tukey's HSD test ($p \leq 0.05$) ($n = 3$).

Element	Leaf 2016			Leaf 2017			Stem 2017			Root 2017			
	PT	PT-DRS	DRS	PT	PT-DRS	DRS	PT	PT-DRS	DRS	PT	PT-DRS	DRS	
Manganese (Mn)	53.7 a	<20.0 b	<20.0 b	23.1 a	30.1 ab	47.4 b	<20.0	<20.0	<20.0	35.0 a	60.6 b	138 c	
Iron (Fe)	101 a	105 a	101 a	125 a	145 ab	180 b	38.9 a	47.9 a	38.7 a	75.4 a	1350 b	3350 c	
Zinc (Zn)	24.0 a	31.2 ab	38.6 b	36.6 a	38.0 ab	57.3 b	31.7 a	56.8 b	124 c	32.9 a	47.4 a	112 b	
Copper (Cu)	<2.50 a	4.04 b	6.78 c	<2.50 a	4.78 b	8.09 c	<2.50 a	4.82 b	4.54 b	2.93 a	10.7 b	27.1 c	
Molybdenum (Mo)	0.179 a	0.464 b	1.14 c	0.465 a	0.866 b	1.14 b	0.108 a	0.292 b	0.327 b	2.00 a	4.03 b	7.69 c	
Lead (Pb)	0.226 a	0.224 a	0.225 a	0.329 a	0.424 a	0.470 a	0.133 a	0.176 a	0.181 a	0.333 a	3.820 b	6.880 c	
Cadmium (Cd)	0.0916 a	0.166 a	0.334 b	<0.0500	<0.0500	0.0634	<0.0500 a	0.104 b	0.0692 b	<0.0500 a	0.378 b	0.679 c	
Nickel (Ni)	0.232 a	0.335 ab	0.469 d	0.368 a	0.597 ab	0.647 b	0.373 a	0.434 a	0.721 b	0.592 a	5.500 b	8.790 c	
Chromium (Cr)	0.241 a	0.318 a	0.283 a	0.501 a	0.558 a	0.898 b	0.456 a	0.509 a	1.16 b	0.518 a	10.6 b	13.4 c	
Mercury (Hg)	0.0362 a	0.0287 a	0.0278 a	0.0263 a	0.0353 a	0.0309 a	<0.0100	<0.0100	<0.0100	<0.0100	0.0189	0.0554	
Cobalt (Co)	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200	<0.200 a	0.704 b	1.81 c
Antimony (Sb)	0.0253 a	0.0173 a	0.0211 a	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500 a	0.0774 b	0.205 c	
Arsenic (As)	0.0922 a	0.202 b	0.207 b	0.0662 a	0.252 b	0.284 b	0.0227 a	0.0536 b	0.0645 b	0.0613 a	0.58 b	1.91 c	
Selenium (Se)	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100 a	0.143 b	0.363 c	
Aluminum (Al)	107 a	112 a	101 a	119 a	220 b	173 ab	49.0 a	50.7 a	122 b	82.9 a	1830 b	4710 c	
Beryllium (Be)	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02 a	0.0476 b	0.197 c	
Tin (Sn)	0.0522 a	0.0596 a	0.109 b	0.451 a	0.340 a	0.405 a	0.255 a	0.265 a	0.288 a	0.349 a	0.468 ab	0.734 b	
Strontium (Sr)	63.1 a	61.6 a	68.5 a	58.2 a	55.8 a	48.0 a	35.4 a	39.4 a	52.2 a	74.2 a	91.4 ab	139 b	
Silver (Ag)	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	
Thallium (Tl)	<0.0200 a	0.0288 a	0.056 b	<0.0200	0.0216	0.0378	<0.0200	0.0226	0.0369	<0.0200 a	0.043 b	0.0941 c	
Vanadium (V)	<0.200	<0.200	<0.200	0.245 a	0.444 b	0.341 ab	<0.200	0.212	<0.200	0.406 a	5.33 b	13.0 c	

3.4. Ions' Content and Pesticide Residues' Determinations in Pomegranate Fruits

Pomegranate fruits were only obtained in the second year of the experiment, since in the first year, the plants were too young to produce fruits. The pomegranate fruit can be divided into two parts: (a) the pulpy seeds, where there is fruit juice, and (b) the peel (rind with the carpellary membranes). Since the peel can also be destined for human and/or animal consumption, it was analysed separately, taking into account the final forms. The results are shown in Table 5. The metal ions' content (Table 5) did not reach the limits

established by the European legislation (1881/2006, 2015/1006, 488/2014 and 2015/1005), by the United Nations Food and Agriculture Organisation (FAO) and the World Health Organisation (WHO) (1995 and later revisions), and other worldwide regulations on heavy metals (Australian, New Zealand, Canada, Brazil and USA).

Table 5. Metal ions' content (mg kg^{-1}) in the pomegranate fruit parts (peel and seed) cultivated on the different substrates (Pt, DRS and Pt-DRS) during the two experimental years (2016 and 2017). Different letters within a column indicate significant differences by Tukey's HSD test ($\rho \leq 0.05$) ($n = 3$).

Element	Seeds			Peel (Rind + Carpillary Membranes)		
	PT	PT-DRS	DRS	PT	PT-DRS	DRS
Molybdenum (Mo)	0.06 a	0.07 a	0.08 a	0.06 a	0.07 a	0.07 a
Tin (Sn)	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20
Cobalt (Co)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Antimony (Sb)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Arsenic (As)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Beryllium (Be)	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Strontium (Sr)	<0.50	<0.50	<0.50	1.13	0.97	<0.50
Total Iron (Fe)	8.05 a	3.39 b	4.17 b	2.87 a	2.94 a	3.01 a
Total Manganese (Mn)	<2.00	<2.00	<2.00	<2.00	<2.00	<2.00
Silver (Ag)	<0.02	<0.025	<0.02	<0.025	<0.02	<0.02
Thallium (Tl)	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Vanadium (V)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Selenium (Se)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Aluminum (Al)	<0.5	<0.5	<0.5	<0.50	<0.50	<0.50
Total copper (Cu)	<0.5 a	0.93 b	1.59 c	<0.50 a	0.63 b	1.23 c
Total Zinc (Zn)	3.43 a	3.22 a	5.58 b	3.40 a	3.18 a	3.35 a
Lead (Pb)	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Cadmium (Cd)	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Nickel (Ni)	<0.10	0.21	<0.10	0.33	0.29	<0.10
Chromium (Cr)	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
Mercury (Hg)	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005

In all the pomegranate edible parts, only Mo, Fe, Zn and Ni were detected above the equipment detection limits. All the metal ions were present in the three substrates, but with differences between them depending on the metallic species. Thus, the Pt-seeds presented the maximal value for total iron (8.05 mg kg^{-1}) and minimal values for total copper and molybdenum ($<0.05 \text{ mg kg}^{-1}$ and 0.06 mg kg^{-1} , respectively), while the Pt-peel presented the lowest content for total iron (2.87 mg kg^{-1}) and total copper ($>0.5 \text{ mg kg}^{-1}$). In contrast, DRS-seeds presented the highest values for molybdenum (0.08 mg kg^{-1}), total copper (1.59 mg kg^{-1}) and total zinc (5.58 mg kg^{-1}). Pt-DRS-peel only showed significant values for total zinc (3.18 mg kg^{-1}). Note that strontium was only detected in pomegranate peel grown in Pt (1.13 mg kg^{-1}) > Pt-DRS (0.97 mg kg^{-1}).

Of the 623 total pesticide residues analysed, only the nitrates were found in both the pomegranate fruit parts. In general, pomegranate seeds, Pt-DRS (11.4 mg kg^{-1}) > Pt (10.7 mg kg^{-1}) > DRS (10.6 mg kg^{-1}), presented higher nitrate concentrations than pomegranate peel, Pt (0.4 mg kg^{-1}) < Pt-DRS (10.6 mg kg^{-1}) < DRS (10.2 mg kg^{-1}). In all cases, the nitrates' concentration obtained was well below the limits established in the European legislation (1881/2006 and 1258/2011), which modifies the former with respect

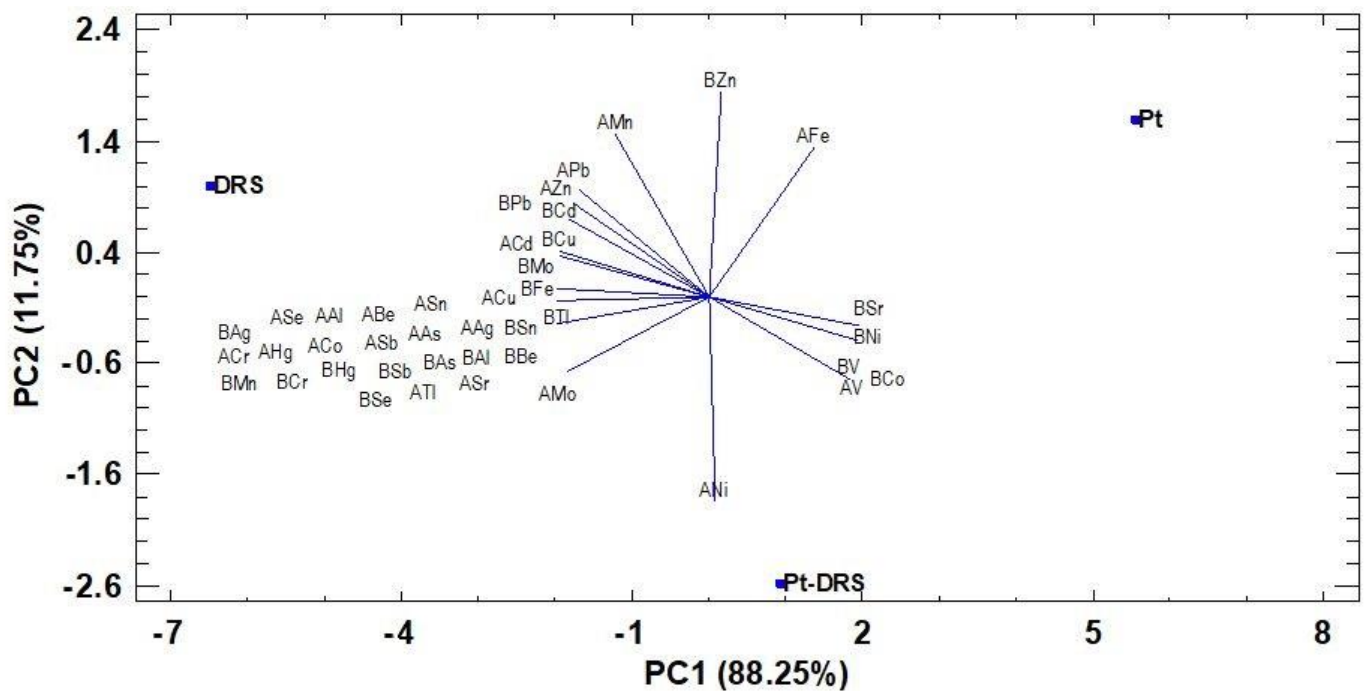


Figure 4. Principal component analysis between the three substrates used (Pt, Pt-DRS, DRS) versus the metal ions analysed in the pomegranate fruits (A: seed, B: peel (rind + membranes)).

5. Discussion

The study began in May 2016, so the first year of growth was from May to December, as opposed to the second year (2017), which lasted from January until September. During the first year, less water was applied, as the plants were smaller and had less requirements. During the first year of cultivation from the time of planting, as the plants were smaller and had less irrigation needs, there were no significant differences in their irrigation needs or drainage of the three substrates.

However, starting in the month of June in the second year of growth and until the end of the assay (September 2017), coinciding with the greatest size of the plants and the greatest rate of evaporation, there were significant differences between the substrate treatments. More water was used with the peat substrate in the months of July and August, which was about 35% greater than with the bioremediated sediment, and the mixed sediment showed water needs that were intermediate between the other two.

The drainage water EC during the first months of cultivation showed some irregularities, but there was a tendency for the peat to have a greater EC, while starting with the second year, this tendency changed, whereby the drainage water from the peat treatment had the smallest EC, and the DRS and Pt-DRS had the greatest EC. This may be due to the heavier texture of the sediment compared to peat (a lighter and more permeable substrate), so the substrates with higher sediment content (pt-DRS and DRS) presented a higher salt-retention capacity and therefore higher EC. However, during the second year, as the irrigation increased, it facilitated the drainage of salts from the sediment, and perhaps those salts that had accumulated in the first months of cultivation were drained as well. In fact, more important EC differences between the substrates were observed in the month of July in the second year of cultivation, coinciding with the time of the greatest contribution of irrigation [33]. In this sense, Kazamias et al. [34] described variable EC values, but these generally increased with the inorganic content of the compost.

The pH variations detected from the beginning to the end of the experiment can confirm the hypothesis of drainage of salts from the sediment described above, as the peat had a significantly lower pH during the first months of cultivation, possibly due to the greater content of bases in the sediment [35]. As the cultivation moved forward, the pH values were similar, probably due to the progressive alkalisation of the peat throughout the

two years of the experiment. A similar behaviour of the pH was also observed by Massa et al. [36] when working on a study of two types of compost as compared to peat.

Between the first and the second cultivation year (2016 and 2017), the DRS drainage showed a decrease in the content of sulphates and nitrates, 4% and 70% respectively, while the ammoniacal nitrogen content increased. For the rest of the analysed minerals (Table 1), their concentrations decreased in the second year with respect to the first, between 28% in the case of dihydrogen phosphate and 49% for boron and molybdate. This decrease was probably due to the absorption by the plant and the washing provoked by the irrigation. The elements that mostly contributed to the salinity were sodium, chloride, potassium, sulphate, nitrate, calcium and magnesium. These results were similar to those found by Jayasinghe et al. [37] in their evaluation of compost as an alternative to peat.

The heavy metals' concentrations in the DRS drained water during the second year decreased by 50% with respect to the content in the first year (Tables 1 and 2), resulting in levels lower than the limits established by the Spanish regulations on the quality of surface water (Spanish Royal Decree 849/1986) and also complying with the demands of the European legislation (2008/105/CE). Jayasinghe et al. [37] have also shown that the total and extractable content of Cu, Zn, Cr, Mn and Pb in different media significantly increased as compared to the peat control, but the content was always below the limits recommended by the United States Environmental Protection Agency [38].

Of the 623 pesticide residues analysed in the DRS drained water, for each of the 2 years of the experiment, 3 pesticides were detected in 2016 and another 3 in 2017. In both cases, the levels were well below the maximum limit authorised by the Spanish (Spanish Royal Decree 849/1986) and European (2008/105/CE) legislation.

It is necessary to know the concentration of heavy metals and minerals in plant material because of their potential toxicity and risks to human health. In this sense, the content of metals in fruits could be an indicator of the contamination level of the soil where the plants were grown [39,40]; therefore, there is a proportional relationship between the increase in the concentrations of extractable Zn and Cu and the increase of their concentrations in plant tissues [41].

When comparing the content of heavy metals between the different parts of the plant (leaves, stem, roots and fruit) (Tables 4 and 5), it was clearly observed that the fruits had the lowest heavy metals' concentration. The concentration of heavy metals and minerals in the different non-edible parts of pomegranates (roots, stems and leaves), as well as in the fruits (seeds, rind and carpellary membranes) did not reach the legal limits that are considered toxic. In the leaves, 8 out of the 21 elements analysed were below the limit of detection, and the rest were below the limits permissible for human consumption (CE 1881/2006, EU 2015/1006, EU 488/2014, EU 2015/1005). Similar conclusions were reached by Jayasinghe et al. [37] in a study on the concentration of heavy metals in broccoli tissues grown in compost.

Zinc is one of the metals that in the present study appeared in high concentrations in plant tissues (edible and non-edible), as was observed by Smith [42] and Fiasconaro et al. [40]. These authors explained that this occurred because Zn is relatively labile, thus it is easily transferred between tissues, and is usually found in greater concentrations in sludge as opposed to other substrates. Zinc is an essential metal and has been shown to be biologically active [43]. The basic function of Zn in plants is related to the metabolism of carbohydrates, proteins, phosphates, auxins, ribosomal structures and for stimulating the resistance of plants in arid climates, as well as the tolerance of bacterial and fungal diseases.

Copper, on the other hand, tends to be strongly absorbed by the soil, and plants regulate the absorption in a more efficient manner as compared to Zn [44], so the concentrations of Cu in the plant, and therefore in the fruit, are usually lower. In this study, the highest concentration of Cu was found in the seeds of the fruits produced on DRS, with a value of 1.59 mg kg⁻¹, which was much lower than those found by other authors in the mesocarp of sweet orange (0.9–3.9 mg kg⁻¹) [45] and banana peel (12 mg kg⁻¹) [46], and the EU

legislation allows up to 10 mg kg⁻¹ in fruits, so the value obtained in our study was six times lower than the maximum limit tolerated by European norms.

Lead is a potentially carcinogenic element in humans, with cultivated plant-based foods being the main route of exposure to this metal [47]. Plants absorb Pb passively, and its translocation to the fruits is difficult. Thus, in the analysis of the fruits, its concentration was below the limits of detection (<0.020 mg kg⁻¹) (EU 2015/1005, which refers to the content of lead in specific food products). These results were in agreement with those obtained by Oliva et al. [39] in sour orange fruits.

Cadmium can accumulate in the human body and produce adverse health effects, and one of the main entry routes into the human body is via food intake. Regulation (EU) 488/2014, establishing the maximum content of Cd in vegetables, regulates the content of cadmium in foods and fruits at 0.050 mg kg⁻¹. In this study, Cd was not detected in the fruits, neither in the seeds nor the rind or carpellary membranes.

For nickel, a carcinogenesis element, concentrations of up to 0.335 mg kg⁻¹ in the skins of the fruit, and concentrations of 0.212 mg kg⁻¹ were obtained in the seeds, while Oliva et al. [39] showed values of Ni in the mesocarp of sour orange within the range of 0.15–1.33 mg kg⁻¹. Markert [48] reported that a normal value of Ni in a reference plant is 1.5 mg kg⁻¹, which indicates that all the values obtained in the present study for fruits can be considered normal. Likewise, the Brazilian legislation on heavy metals states an upper limit of up to 5 mg kg⁻¹.

Iron and manganese are essential elements for plants and are involved in many enzymatic systems, and therefore they are found in all plants in variable quantities, depending on species and plant parts analysed. The content of Fe in the rind and the carpellary membrane were not found to be significantly different between the fruits produced with the sediment and those produced with commercial peat (control substrate). However, the content of Fe in the seeds was double in fruits produced with peat as compared to fruits produced with sediment (Table 5). This could be explained by the greater acidity of the peat, which at the same time results in a greater availability of Fe²⁺ for the plants. The content of Mn in the pomegranates grown with peat (Pt), the mixture (Pt-DRS) and sediment (DRS) was below the limits of detection used in this study (<2.00 mg kg⁻¹), as well as the rest of the metals shown in Table 5 (Sn, Co, Sb, As, Be, Mn, Ag, Tl, V, Se, Al, Pb, Cd, Cr and Hg). In this sense, it can overall confirm that the fruits obtained with the bioremediated sediment assayed in this study did not contain heavy metals, and those that were present were found below the limits indicated by the European legislation.

Based on the potential risk of residue accumulation in the fruits, due to the contaminants' absorption by the roots, which are in direct contact with the sediment, a pesticide residues' scan in the different pomegranate fruit parts (seed and peel plus carpellary membranes) was also carried out. In total, 623 pesticide residues and other contaminants were analysed, with only nitrates detected in the seeds (between 10.6 and 11.4 mg kg⁻¹), as well as in the rind and membranes (between 10.2 and 10.6 mg kg⁻¹). In both parts of the fruit, the nitrate concentration obtained was below that established in Regulation (EU) 1258/2011 of the Commission from 2 December 2011, for the most restrictive case (infant food for lactating children and young children, <200 mg NO₃⁻ kg⁻¹). Thus, from the point of view of the presence of pesticide residues, the fruits produced with the use of this sediment were free from contamination, so the fruits are apt to be consumed fresh as well as after their transformation.

Based on the interpretation of principal component analysis results, along with the data presented in Tables 4 and 5, it can be concluded that for the fruits (seeds, as well as rind and carpellary membranes), the concentrations of heavy metals and other metals were below the maximum limits allowed by different laws. In general, the highest heavy metals' concentrations were found in plants grown in DRS.

Considering the results obtained in this study, the European Commission should consider a change in legislation that would allow the use of this resource (dredged remedi-

ated sediments) as a source of new substrates for agricultural use, in order to reduce the environmental and economic problems created by these sediments.

6. Conclusions

The results obtained in this study confirm the potential of DRS for its use as an agricultural substrate, alone or mixed with other substrates, since the trees presented an adequate and satisfactory development. In addition, in the pomegranate fruits obtained with the sediment as culture media, only Mo, Fe, Zn and Ni were detected, but in all cases with values below the European legal limit, hence pomegranate fruits could be considered suitable for human consumption.

From a global environmental point of view, the results confirmed that the reuse of DRS in agriculture would not generate a significant pollution impact since all the pollutants monitored (metal ions and pesticide residues), both in the sediment and in the DRS drainage, presented values below the specific limits established both in Spanish and in European legislation.

As a global conclusion, it can be highlighted that the results verified the suitability of this sediment as a growing substrate in the nursery, as well as for the growing of trees in pots, which could diminish the pressure for exploiting natural substrates such as peat. Besides, from this study, it was possible to verify an environmentally sustainable use potential of dredged marine sediments of ports all over the world, thereby contributing to mitigate the environmental impact.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/su13179661/s1>, Figure S1: Pictures of pomegranate trees used in this study. Table S1: Initial characteristics of remediated dredged sediment used in this study. Values are mean \pm standard deviation ($n = 3$). Adapted from Tozzi et al., 2020.

Author Contributions: Conceptualisation, methodology and validation, formal analysis, investigation, writing—original draft preparation, writing—review and editing, J.J.M.-N., P.L., F.H., R.M.-F., E.G. and P.M.; supervision, P.M.; project administration and funding acquisition, P.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the European LIFE Project, HORTISED, grant number LIFE14/ENV/IT/000113.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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