Recycling agricultural, municipal and industrial pollutant wastes into fertilizers for a sustainable healthy food production

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Abstract

This work was focused on recycling different typology of pollutant wastes (olive pomace and orange residues; municipal wastes and sulphur residue of hydrocarbon refining processes) with the triple objectives of limiting wastes in landfill, reducing greenhouse gas emission and producing organic-mineral fertilizers. The environmental risk and benefit of the whole process have been considered. The specific objectives were: 1) innovation in waste management techniques by reducing the accumulation of different typology of wastes with a unique process 2) verifying efficiency of the obtained organic-mineral fertilizers on soil and plant growth 3) improving soil and crop quality connecting waste and food, to economy and environment.

Sulphur-based pads increased soil quality and the best soil improvement was observed when the pads contained orange residues. Onion and Garlic showed the best growth when cultivated in presence of sulphur-based pads (+ 20%), the best performance was observed when orange residues were present in the pads (+45%). Onion and Garlic quality in terms of antioxidant compounds and antioxidant capacity increased in presence of sulphur-based pads (+ 30%) mostly when orange residues were present in the pads (+90%). In short, in addition to the environmental advantages, numerous economic benefits coming from the decrease in the production and use of chemical fertilizers, the reduction of costs for landfilling and the gain rising from the sale of the new fertilizers produced, emerged.

Keywords: crop quality; mineral-organic fertilizer; soil fertility; sulphur bentonite; waste management.

1. Introduction

In the last 50 years, the green revolution using genetically selected plant varieties, fertilizers, pesticides, water and other capital investments, allowed a significant increase in agricultural production (more than three times) and population worldwide (FAO, 2017a; FAO and OECD, 2019). The green revolution (with a production growth of 23.7 million food tons per day) was criticized, because caused biodiversity loss, dependence from fossil biofuels and pollution impacting negatively soil, air and water resources (FAO, 2017b), putting at risk population health and ecosystem sustainability.

Agriculture is the sector that generates about one fifth of greenhouse gas emissions worldwide but, at the same time, produces a large amount of biomass (European Commission, 2015). The latter could represent an essential environmental and bioeconomic intake (Bracco et al., 2018; European Commission, 2012), because biomass utilization can reduce dependence from fossil fuel and consequently mitigate greenhouse gas emissions (McCormick and Kautto, 2013). The transformation of vegetable wastes into products with added value, can contribute to the evolution of new green markets worldwide.

In addition to the agricultural sector, the industrial sector produces also wastes that could be recycled in valuable products valorising waste chain. In the refining process of crude oil, the excess of sulphur, a compound essential for life with many industrial applications, it's unwanted and needs to be removed for its damaging effects not only on the environment, but also on catalytic cracking and refining processes due to the corrosive effects, and the generation of acid gasses during the combustion (Al-Bidry and Azeez, 2020). More than 90% of the elemental sulphur recovered is used to produce sulphuric acid, even if its production generates emissions which are harmful for the environment. Marwa et al. (2017), with a life cycle assessment study, showed that sulphuric acid production system not only impacts the environment with high CO_2 emission (83.26 kg/ ton of sulphuric acid) but also it is energetically unsustainable. In agriculture the value of sulphur is well known for more than a century (Bogdanov, 1899; Hart and Peterson, 1911), however, the incessant utilize of other formulations containing nitrogen (N) and phosphorus (P) but no sulphur (S), the higher S exportation from soil under high yield crops, and the decreased S input by rainwater, led to an increment of S deficiency in soils (Lucheta and Lambais, 2012). In the last years, waste materials raised the attention of sector operators and politicians in view of circular economy because their use in agriculture reduces the waste of valuable nutrients, keeping them into the ecosystem. Waste materials, with their organic and elemental contents, can ameliorate soil properties promoting in turn crop performance (Al-Barakah et al., 2013; Song et al., 2015) and reducing, at the same time, the use of chemical fertilisers. Previous works evidenced the feasibility of using composted municipal wastes as fertilizer in agriculture (Srivastava et al., 2016) evidencing also that the composition and application rate greatly affected soil microbial biomass. Gelsomino et al. (2010) evidenced that the agricultural wastes (orange and olive residues) were mostly used composted for agricultural purposes, with satisfactory results. On the basis of the above considerations and of our previous works (Muscolo et al., 2017; 2019; Panuccio et al., 2016; 2019) focused on the reuse of different kinds of biomasses for land restoration and crop improvement, the novelty of this work is to recycle raw material crude and not composted with low cost, low emission and high incoming, sulphur, municipal waste and polluting agricultural wastes of local origin: orange peel and pulp, commonly called "pastazzo, and olive pomace with a unique process, to produce fertilizers able to recovery soils and improve crop quality and yield. The aim is the development and setting of a new market of mineral-organic fertilizers. The new fertilizers will be experimentally produced by Steel Belt System s.r.l, with a patented technology that already uses Sulphur finely mixed with bentonite clay to make it friable and easily absorbable by plants. The aim of this work was to contribute to address environmental issues such as limiting the excess of sulphur, the wastes in landfill, and the use of chemical fertilizers producing at the same time mineral-organic fertilizers which are not polluting for soil and water. To close the loop in a sustainable and productive way the specific objectives were: 1) innovation in waste management techniques by reducing the accumulation of different typologies of wastes through a unique process, using not composted agricultural wastes 2) verifying efficiency of the obtained mineral-organic fertilizers on soil and plant growth 3) improving soil and crop quality connecting waste and food, to economy and environment. For this purpose, raw materials have been analysed from a chemical, physical and biological point of view, and the composition of the mineral-organic fertilizers was optimized starting from previous works (Muscolo et al., 2019). Sulphur, insoluble in its elemental form, when mixed with bentonite-clay and wastes is slowly released into soil, where bacteria transform it in sulphate, the chemical form soluble in soil and easily absorbed by crops. Additionally, the organic wastes (agricultural and municipal) used in dried form and mixed to sulphur and bentonite add organic components to soil maintaining soil biodiversity equilibrium.

2. Material and Methods

2.1 Fertilizer preparation

The manufacturing process to obtain pads of sulphur with a diameter of 3/4 mm was carried out by Steel Belt System s.r.l. as reported in Muscolo et al. (2017; 2019). Sulphur was linked to bentonite clay (as support and carrier). The amount of bentonite used proportionally to molten S is based on an arbitrary 10%. To prepare the different pads sulphur-bentonite was mixed with orange residue (pastazzo) (Or), or olive pomace (Op) or dried municipal waste (Mw) sieved at (0.2-0.1 mm), or with a mix of them. Elemental S was in percentage the main constituent of the pads.

1. Preparation phase: 90% elemental S was pelletized with 10% bentonite clay (as support and carrier). These pads represent our control. 85% elemental S was pelletized with 10% bentonite clay and with 5% orange waste, 5% olive pomace or 5% municipal waste.

2. Pelletized phase: once prepared the mixture of liquid S with the ingredients (bentonite and/or agricultural or municipal wastes), the obtained mixtures have been introduced in a special patented rotary pastillator, which deposits the liquid pads of the above listed ingredients opportunely mixed, on a heat exchanger in continuous steel tape for the solidification of the pads.

Pathogens (total coliforms, faecal coliforms, salmonella spp and Escherichia coli) and heavy metals have been also assessed to avoid any toxic and harmful effects on soils and crops. Samples for metals were preserved with nitric acid and then analyzed by atomic absorption spectroscopy (GBC mod. 908). Total coliforms, faecal coliforms and escherichia coli were expressed as densities of colonies $\log_{10}$ CFU 100 g^{-1} waste material. The same samples were also analyzed for Salmonella spp., according to a procedure consisting of a ‘pre-enrichment’ stage using a buffered peptone water solution and a non-selective culture medium to revitalize the microorganism as reported in Ben Said et al. (2017).

2.2 Soil treatments

In this experiment a sandy-loam (11.85% clay, 23.21% silt, and 64.94% sand) soil was used (FAO, 1999). The experiment was performed using pots of 30 cm diameter each containing 9 kg of soil with a pH of 8.87, 1.81 % of organic matter. Pots were amended with S-bentonite (SB); S-bentonite + orange waste (SBOr); S-bentonite + olive pomace (SBOp), S-bentonite + municipal wastes (SBMw), S-bentonite + orange waste+ municipal wastes (SBOrMw) and S-bentonite + olive pomace + municipal wastes (SBOpMw) at the concentration of 1.4 g corresponding to 476 kg S ha^{-1} dose generally used to lower the pH and to replenish S (Severson and Shacklette, 1988; Muscolo et al., 2017). Non-fertilized soil was used as control (CTR). The experiments were performed in triplicates in greenhouse as reported in (Muscolo et al., 2017). During the experiment, the pots were watered regularly to ensure that water content was maintained at 70% of field capacity. At the end of the experiments (90 days after treatments) the differently treated soils (three replicates), were air-dried and sieved (<2mm) prior to the chemical analysis (fully described in the section soil and pad analysis). Soil samples for the biochemical determination (microbial biomass and enzyme activities) were stored in the refrigerator at 4°C for up to 24 h until processing.

2.3 Soil and pad analysis

Electric conductibility (EC) was determined in distilled water by using 1:5 residue/water suspension, mechanically shaken at 15 rpm for 1 h to dissolve soluble salts and then detected by Hanna instrument conductivity meter; pH was measured in distilled water (soil/pad:solution ratio 1:2.5) with a glass electrode. Organic carbon was assessed with dichromate oxidation method (Walkley and Black, 1934). Total nitrogen (TN) was measured with Kjeldahl method (1883). C/N was determined as a carbon:nitrogen ratio. Microbial biomass carbon (MBC) was determined in field moist samples (equivalent to 20 g D.W.) (Vance et al., 1987). Soil extracts of both fumigated and unfumigated samples were filtered and analysed for soluble organic C (Walkley and Black, 1934). MBC was estimated on the basis of the differences between the organic C extracted from the fumigated soil and that from the unfumigated soil, and an extraction efficiency coefficient of 0.38 was used to convert soluble C into biomass C (Vance et al., 1987).

Water soluble phenols were extracted in triplicate as reported by Kaminsky and Muller (1977; 1978). Total water-soluble phenols (monomeric and polyphenols) were determined by using the Folin-Ciocalteau reagent (Box, 1983). Tannic acid was used as a standard and the concentration of water-soluble phenolic compounds was expressed as tannic acid equivalents (µg TAE g^{-1} D.W.).

Fluorescein diacetate hydrolase (FDA) was determined according to the method of Adam and Duncan (2001). Dehydrogenase (DHA) activity was determined by the method of von Mersi and Schinner (1991). Cations and anions were detected by ion chromatography (DIONEX ICS-1100). For anions, 0.5 g of dried material was extracted using 50 ml of anion solution ($Na_2CO_3/NaHCO_3$ 3.5 mM) stirring for 20 minutes. The extracts have been filtered and the chromatographic analysis was carried out. For cations, 1 g of dry material was ashed at 550 °C for 5-6 hours in a porcelain capsule. The ash was then mineralized for 30 minutes at 100 °C using 1M HCl solution. The solution was subsequently filtered and analysed by ion chromatograph (eluent meta-sulfonic acid 20 mM).

2.4 Plant analysis

In the present investigation the sulphur-based fertilizers have been tested on *Allium cepa* L. (the common onion) and *Allium sativum* L. (the common garlic), sulphur loving crops, that contain important substances with protective and beneficial effects on human health. The presence of sulphur-containing phytochemicals in garlic and onion provides substantial immunomodulatory, anti-inflammatory, anticancer, antitumor, antidiabetic, anti-atherosclerotic, and cardioprotective features. The experiment was terminated at bulb maturity, as characterized by neck softening and reduced solution uptake. Bulb diameters were measured using callipers and leaves were counted. Leaf and root length were measured with a meter. Plants were harvested and separated into shoots, bulbs, and roots. Fresh weights were measured by weighing, and the individual plant parts were then dried at 70 °C in an oven. Dry weights were determined and plant materials were ground to pass a 20-mesh screen. Antioxidant compounds and antioxidant activities have been detected in the onion bulbs differently fertilized in comparison to control at the end of the experiments. Antioxidant and antioxidants activity are markers of crop quality because related to the beneficial human health effects (Younes et al., 2021)

2.5 Determination of total phenolic compounds and total flavonoids in plants

Total phenol content, was detected by Folin–Ciocalteu assay (Muscolo et al., 2020). The absorbance of the samples was recorded at 760 nm. A calibration curve was constructed with gallic acid and results were expressed as g gallic acid equivalent kg^{-1} DW. Total flavonoids in the extracts were detected according to the spectrophotometric method (Muscolo et al., 2020). The absorbance was measured at 430 nm. Flavonoid content was calculated from a calibration curve of rutin and expressed as g rutin equivalent kg^{-1} DW.

2.6 Determination of antioxidant activities in plants

The antioxidant activity against DPPH radical (2,2-diphenyl-1-picryl-hydrazyl-hydrate) was determined according to Muscolo et al. (2020). The DPPH concentration in the cuvette was chosen to give absorbance values of ∼1.0. Changes in absorbance of the violet solution were recorded at 517 nm after 30min of incubation at 37 °C. The inhibition I (%) of radical-scavenging activity was calculated as

$$I\ (\%) = [(A0 - AS)/A0] \times 100,$$

where A0 is the absorbance of the control and AS is the absorbance of the sample after 30 min of incubation. Results were expressed as Trolox equivalent (TE).

The ABTS assay was performed according to Muscolo et al. (2020). The absorbance of the samples was recorded at 734 nm using a UV–visible spectrophotometer. The inhibition I (%) of radical-scavenging activity was calculated as $I\ (\%) = [(A0 - AS)/A0]\times100$, where A0 is the absorbance of the control and AS is the absorbance of the sample after 4min of incubation. Results were expressed as μmol L^{-1} TE using a Trolox (1–50 μmol L^{-1}) calibration curve.

The oxygen radical absorbance capacity (ORAC) assay was performed according to Muscolo et al. (2020). ORAC values were expressed as μmol TE mg^{-1} FW using a Trolox (10–100 μmol L^{-1}) calibration curve.

2.7 Statistical analysis

Analysis of variance was carried out for all the data sets. One-way ANOVA with Tukey's Honestly. Significant Difference test were carried out to analyse the effects of fertilizers on each of the various parameters measured. ANOVA and T-test were carried out using SPSS software (IBM Corp.2012). Effects were significant at $p \leq 0.05$.

3. Results and discussion

3.1 Chemical properties of biomass

The biomass used to prepare the different sulphur-based pads, differed in numerous chemical parameters, SBMw was alkaline in respect to olive pomace and orange residue and had the lowest EC and moisture content than Or and Op. Organic carbon and total nitrogen were the highest in Op (57%), and significantly the lowest in SBMw (6.3%), (Table 1). Nevertheless, the low amount of carbon, nitrogen, ammonium and potassium, SBMw contained high amount of calcium, magnesium and sulphate (Table 1). All the biomass analysed contained important macro and micro elements useful for mineral plant nutrition and were eligible to be used for soil fertilization purpose. Municipal wastes have been largely used in developing countries to produce compost an attractive procedure to dispose this waste reducing the huge problem of landfill conferment. Generally, the Mw was used composted or dried (De Bertoldi et al., 1996; Zinati et al., 2004; Kabirinejad and Hoodaji, 2012) to reduce pathogens even if other two drawbacks, such as excess of soluble salt accumulation and potential toxicity of certain elements to plants, were identified (Maftoun et al., 2004). It is well-known that micro-pollutants, present in these wastes, can cause adverse effects on organisms and can modify soil properties (Muir and Howard, 2006; Carbonell et al., 2009). Composting Mw for agricultural purposes is a way to break down pathogen organisms but not its content of heavy metal that could pollute soils (Weber et al., 2007) and lower plant productivity and quality with detrimental effects on human health (Ashfaque et al., 2016). Our results evidenced that while Or and Op didn’t contain heavy metals, the Mw that we used in this research, contained cadmium, lead, zinc, nickel, mercury, copper and chromium but their amounts fell in the range allowed by European regulation and were far below the allowed limit (Fig. 1). On the basis of these analyses, from which resulted that the biomasses were suitable for the environment, Steel Belt System used them to produce fertilizers in the form of round shape pads with small diameter to favour the fast release of nutrients and sulphur in soil. No pathogens (total Coliforms, faecal Coliforms, E. coli and Salmonella spp), have been detected in the pads. The process used for pads production destroyed all the living forms that potentially can be found in municipal wastes.

Chemical soil analysis, 3 months after the addition of the different fertilizers, evidenced significant and substantial differences among the treatments, and between the treatments and the control. Water content increased in all the amended soils, except for SB, suggesting that the addition of fertilizers with organic components increased water holding soil capacity (Vengadaramana & Jashothan, 2012; Mirzabaiki et al., 2020). No significant differences were detected in pH values measured in H_2O and KCl between control and treatments, and among the treatments (Table 2). EC increased in all the amended soils and mostly in presence of the mixed pads, SBOrMw and SBOpMw (Table 2), that originally were richer in chemical elements. EC was in any case in all the soil samples, lower than 4 dS/m, the threshold for which a soil is considered saline.

Water soluble phenols increased only in soils fertilized with SBOp, suggesting that during the three months of treatment, the phenols contained in the Op raw material, have been released into the soils. This data evidences how the composition of raw material is able to affect some soil properties. Organic carbon enhanced in all treatments compared to control (CTR), except for SB. The greatest increase was observed in soils treated with SBOr and SBOrMw. The lowest value was observed in SB treatment. Data evidenced that the addition of pads to soils containing orange residues increased the amount of carbon in soils. C/N ratio varied with the treatments and it was the highest in SB, suggesting that, with this treatment, the process of organic matter decomposition tended to remain fairly stable over the 3 months. FDA increased only in SBOr and SBOp. The lowest value was observed in CTR, that had also the lowest amount of microbial biomass and the lowest dehydrogenase activity (Table 2). Fluorescein diacetate hydrolase reflects the potential microbial activity of soil freshly amended with a wide range of organic material, and generally increases when the microbial activity increase. This increase is inversely related to the degree of stabilisation of the added organic matter, defined by the C/N ratio (Sánchez-Monedero et al., 2008). Our results agree with the above findings, showing a strict positive relationship between FDA and MBC, and an inverse correlation between FDA and C/N. MBC and DHA had the same trend of FDA were the lowest in CTR and SB, and increased with pads containing organic wastes and mostly with pads containing agricultural wastes. The ions in soil changed with the type of fertilizer added, a significant increase in calcium, magnesium and sulphate was observed in soil amended with fertilizers containing orange residues and Mw, the wastes which, already initially, contained these nutrients in major amount (Fig. 2). These data agree with data of Hussain et al. (2017), showing that organic wastes had a great positive impact on soil properties including the addition of nutrients.

3.2 Plant growth and antioxidant properties.

Regarding onion plants, the only increase in leaf length, respect to control, was observed when plants were grown with pads containing orange residue (Fig. 3). Root length significantly increased mostly in presence of SBOr, and at minor extent with SB, and SBOrMw (Fig. 3). Bulb diameter increased in all the treatments except for CTR and SBOpMw (Fig. 3). Leaves were less numerous in SB, CTR and SBOpMw compared to the other treatments (Fig. 3). Pads containing orange residue showed the best physiologic effect on plants. Pads with orange had the greatest amount of ammonium, potassium and a good amount of magnesium and calcium. Previous studies of Backes et al. (2018) and Nawaz et al. (2017) demonstrated the important effect of nutrients on onion growth and development, putting in evidence as bulb diameter increased in presence of potassium and ammonium irrespectively of plant growth. Our results agree with findings of the previous authors and with results of Fawzy et al. (2007) which showed as calcium soil application significantly increased vegetative growth and bulb yield.

In this study, total phenols increased in all treatments compared to control and SBOr and SBOrMw were the conditions that better stimulated their synthesis. Surprisingly, SBOpMw increased total phenol content more than SB, SBOp, SBMw, suggesting a synergistic effect of Op and Mw when they were mixed in the pads (Table 3). Total phenols are important antioxidants with beneficial effects on human health. It is well known that total phenol synthesis in plant is highly in competition with protein synthesis, which are indispensable for growth. In environment rich of nutrients, primary metabolism, strictly linked to growth processes, prevails on secondary metabolism. Stefanelli et al. (2010), highlighted that nitrogen fertilization caused a decrease in the quantity of total phenols. Our data evidenced that SBOr and SBOrMw, the pads with a minor content of nitrogen mostly increased total phenol amounts in onion bulb. Similar behaviour was observed for flavonoids. DPPH that measures the scavenger capacity of a plant, increased in presence of SBOr and SBOrMw more than in the other treatments (Table 3) and this increase was correlated to the amount of total phenols. Benkeblia (2005), in his study on garlic and different varieties of onions, demonstrated high significant correlations between total phenolic content and reducing power, scavenging of hydrogen peroxide and chain-breaking activity of extracts. ORAC that measures inhibition of peroxyl radical induced oxidations by antioxidants and thus reflects classical radical chain-breaking antioxidant activity was higher in SBOr and SBOrMw than the other treatments followed by SBOpMw, evidencing one more time the correlation between total phenols and antioxidant activities and between the content of total phenols and the chemical composition of the pads. ABTS, increased in treated onions in respect to control, showing positive relationship with total phenols and flavonoids. These results indicated promising perspectives for the exploitation of onion, and this study could be useful to consumers, planning rich antioxidant diets and to nutritionists in estimating the daily intakes of phenolic antioxidants and their impact on health.

Garlic grew better with treatments than control, the best leaf elongation and leaf number were detected with pads containing orange residues (Fig. 3). Root length decreased only in presence of SB and SBOrMw and SBOpMw, while bulb diameter increased in all treatments compared to control and much more when in the pads were present orange residues (Fig. 3). As in onion, total phenols and flavonoids increased in treated garlics and mostly in presence of orange residues (Table 4). Accordingly, increased also the antioxidants capacities expressed as DPPH, ORAC and ABTS (Table

4) confirming a strict correlation between the amount of total phenols and flavonoids and antioxidant properties of the plants.

3.3 Environmental impact: risk and benefit

By calculating the environmental and economic impact of reusing recalcitrant agro-industrial wastes and putting all the results on the scales, the production of sulphur-bentonite fertilizers can be considered a beneficial process leading to significant reductions in greenhouse gas emissions in the atmosphere for the elimination of a large amount of hazardous materials from the environment. From a review of the literature it emerged that one ton of wet orange waste left on the ground emits 0.130 kg of CH_4, 30.900 kg of CO_2 and 0.069 kg of N_2O (Manfredi et al., 2009) as well as one ton of wet olive pomace produces, if left not treated on the earth's surface, 1162.3 kg of CO_2, 122 kg of CH_4 and 0.12 kg of N_2O. With appropriate recycling of these agricultural wastes by using low cost and efficient processes it is possible to slow down soil and air pollution (Hischier et al., 2020). Considering that pads, in addition to agricultural wastes, contain also recalcitrant sulphur and municipal wastes, the GHG emissions should to be definitively reduced by their absence in dump. Study of Lee et al. (2017) showed an emission of 2603 to 2708 t CO_2e/dry t, from municipal waste abandoned in landfill. CH_4 emitted by landfill was at about 54 kg/ dry t, with a greenhouse warming potential 25 times higher than CO_2. Nitrous oxide is produced predominantly by microbial processes as a by-product of nitrification and as a product of incomplete denitrification, one tonne of nitrous oxide would generate 265 times the amount of warming as one tonne of CO_2. Rinne et al. (2005) and Harborth et al. (2013), sowed a higher emission of nitrous oxide (approx. 0.03–0.4 ml m^{-2} min^{-1}) from waste landfills than agricultural and forest soils (Rinne et al., 2005). LCA modelling performed by Damgaard, et al. (2011) and Manfredi et al. (2011) showed that landfills are the main contributors for global warming, photochemical stratospheric ozone formation. In addition to landfilling wastes, sulphur as residue of hydrocarbon refining processes generates hydrogen sulphide and sulphur oxide causing environmental pollution, thus the production of the pads containing a high sulphur percentage can help to maintain a clean environment. As reported in Pergola et al. (2020) the production of 1 ton of compost on-farm of raw materials caused an energy requirement ranging from 1500 to 2000 MJ, and a mean cost of 130 euro, evidencing that the production cost was in any case cheaper than commercial compost. Our process, which used non-composted organic material, completely reduces these costs, making it more sustainable from both an environmental and energy point of view.

Furthermore, as reported by Haitao et al. (2015) the advantage due to the replacement of chemical fertilizers with organic-mineral ones which leads to -20% GHG with a simultaneous increase (+ 50%) of the soil organic matter must also be considered. In addition to the environmental advantages, the economic benefits can come from the sale of the new fertilizers produced. A ton of pads can be sold on average for 30 euros in EU countries, to which must be added the approximately euros saved by the reduction of CO_2 and CH_4 emissions, the decrease in the production and use of chemical fertilizers and the reduction of costs for landfilling, which allows the manufacturing process to be included as a clean process.

4. Conclusions

This study is an innovative approach of green remediation which combines the recovery of municipal, industrial and agricultural wastes reducing their negative impact on the environment while transforming them in a resource toward achieving circular economy. Results demonstrated an increase in soil and crop quality when Sulphur-based pads were used. The best increase was detected when pads contained orange residues were used. The agricultural utilization of these wastes could meet the target objective of European Union countries to decrease the quantity of wastes going to landfill sites by 20% by 2010 and by 50% by 2050. In respect to other previous studies, it points out the importance that the chemical characteristics of the wastes have on the properties and potential added value of the final products. Data showed many differences between the properties and effectively of the different fertilizers which in some case can overlap, and in other can act in different way. For this reason, their actions and properties need to be discriminated for increasing the efficacity of their use.

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 1 Chemical properties of agricultural (olive pomace and orange residue) and municipal wastes. Organic carbon (OC), total nitrogen (TN), carbon nitrogen ratio (C/N), ions and water soluble phenols (WSP).

Chemical properties	**Olive pomace**	**Orange residue**	**Municipal waste**
pH	5.0^{b*} ±0.1	5.1^{b} ±0.2	7.7^{a} ±0.2
EC (mS/cm)	12.0^{a} ±1.1	10.1^{b} ±0.9	5.0^{c} ±0.2
Moisture (%)	86.7^{a} ±3.2	83.6^{a} ±2.9	55.5^{b} ±2.5
OC (%)	57.6^{a} ±1.9	45.6^{b} ±2.5	6.3^{c} ±1.5
TN (%)	2.0^{a} ±0.6	1.2^{b} ±0.3	0.7^{c} ±0.5
C/N	28.2^{b} ±1.9	36.8^{a} ±1.7	9^{c} ±1.1
Na^{+} (mg g^{-1} dw)	1.8^{a} ±0.5	0.97^{b} ±0.2	0.86^{c} ±0.4
NH_4^{+} (mg g^{-1} dw)	0.24^{b} ±0.03	0.33^{a} ±0.04	0.17^{c} ±0.04
K^{+}(mg g^{-1} dw)	39.2^{b} ±2.3	49.2^{a} ±2.6	6.7^{c} ±0.9
Mg^{2+} (mg g^{-1} dw)	2.2^{c} ±0.4	4.2^{b} ±0.7	12.1^{a} ±0.6
Ca^{2+} (mg g^{-1} dw)	2.5^{c} ±0.7	9.3^{b} ±1.0	60^{a} ±1.6
Cl^{-} (mg g^{-1} dw)	3.8^{a} ±0.5	2.4^{b} ±0.6	1.65^{c} ±0.3
PO_4^{3-}g g^{-1} dw)	2.1^{a} ±0.4	1.1^{c} ±0.3	1.2^{b} ±0.4
SO_4^{2-}(mg g^{-1} dw)	nd	nd	30 ±1.6
WSP (mg TAEg^{-1} dw)	1.8^{a} ±0.4	0.53^{c} ±0.2	1.2^{b} ±0.8

Data are the mean of three independent experiments ± standard errors. *Different letters, in the same row, indicate significant differences at $p \leq 0.05$.

Table 2 Soil properties 3 months after the treatment with the different fertilizers. CTR= Control, soil without fertilizer; soil+sulphur+bentonite (SB); Sulphur+bentonite+orange residue (SBOr); Sulphur+bentonite+olive pomace (SBOp); Sulphur+bentonite+municipal waste (SBMw); Sulphur+bentonite+orange residue+municipal waste (SBOrMw); Sulphur+bentonite+olive pomace+municipal waste (SBOpMw). Water content (WC), electric conductivity (EC, µS/cm), organic carbon (OC), total nitrogen (TN), carbon nitrogen ratio (C/N), water soluble phenols (WSP, µg TAE g^{-1} ds), fluorescein hydrolase (FDA, µg fluorescein g^{-1} ds), Dehydrogenase (DHA, µg INTF g^{-1} ds h^{-1}), Microbial Biomass C (MBC, mg C g^{-1} s).

	CTR	**SB**	**SBOr**	**SBOp**	**SBMw**	**SBOrMw**	**SBOpMw**
WC (%)	10.0^{*c} ±0.2	10.2^{c} ±0.2	13.93^{a} ±0.4	12.5^{b} ±0.1	14.6^{a} ±0.7	14.5^{a} ±0.7	12.88^{b} ±0.4
pH (H_2O)	8.87^{a} ±0.1	8.71^{a} ±0.2	8.72^{a} ±0.1	8.84^{a} ±0.1	8.81^{a} ±0.1	8.63^{a} ±0.2	8.66^{a} ±0.2
pH (KCl)	8.31^{a} ±0.2	8.32^{a} ±0.1	8.25^{a} ±0.2	8.42^{a} ±0.1	8.21^{a} ±0.1	8.05^{a} ±0.3	8.11^{a} ±0.3
EC	352^{c} ±4.1	273.8^{e} ±2.0	382.5^{b} ±2.8	332.2^{d} ±2.2	386.3^{b} ±3.1	466.4^{a} ±7.3	451.1^{a} ±8.1
WSP	2.51^{b} ±0.1	2.52^{b} ±0.1	2.55^{b} ±0.3	3.81^{a} ±0.2	2.57^{b} ±0.4	2.52^{b} ±0.1	2.51^{b} ±0.2
OC (%)	1.047^{c} ±0.02	0.997^{d} ±0.01	1.320^{a} ±0.2	1.197^{b} ±0.05	0.984^{d} ±0.02	1.297^{a} ±0.1	1.140^{b} ±0.04
TN (%)	0.058^{b} ±0.01	0.035^{c} ±0.01	0.084^{a} ±0.01	0.077^{b} ±0.01	0.082^{a} ±0.01	0.081^{a} ±0.01	0.079^{b} ±0.01
C/N	18^{b} ±0.5	28^{a} ±0.9	16^{c} ±0.2	16^{c} ±0.1	12^{e} ±0.1	16^{c} ±0.3	14^{d} ±0.4
FDA	4.25^{e} ±0.2	6.43^{c} ±0.2	8.61^{a} ±0.4	8.20^{a} ±0.3	5.54^{d} ±0.1	7.12^{b} ±0.2	6.52^{c} ±0.2
DHA	49^{e} ±0.8	54^{d} ±1.2	69^{a} ±0.6	65^{b} ±1.2	60^{c} ±1.1	67^{b} ±0.9	56^{d} ±1.0
MBC	1.81^{f} ±0.1	3.55^{e} ±0.1	5.67^{a} ±0.2	5.33^{b} ±0.1	4.44^{c} ±0.2	5.44^{ab} ±0.3	3.88^{d} ±0.1

Data are the mean of three independent experiments ± standard errors. *Different letters, in the same row, indicate significant differences at $p \leq 0.05$.

Table 3 Antioxidant activities (DPPH, µM Trolox Eq/g FW; ABTS, µM Trolox Eq/FW; and ORAC, µM Trolox Eq/100g FW) polyphenols (mg Tr/g FW) and flavonoids (mg rutin/g FW) in red onion bulbs grown for 3 months in soils differently treated: control CTR, soil without fertilizer; soil+sulphur+bentonite (SB); Sulphur+bentonite+orange residue (SBOr); Sulphur+bentonite+olive pomace (SBOp); Sulphur+bentonite+ municipal waste (SBMw); Sulphur+bentonite+orange residue+ municipal waste (SBOrMw); Sulphur+bentonite+olive pomace+ municipal waste (SBOpMw).

Onion	**CTR**	**SB**	**SBOr**	**SBOp**	**SBMw**	**SBOrMw**	**SBOpMw**
DPPH	2.81*^{d} ±0.04	3.13^{c} ±0.05	4.04^{a} ±0.18	3.05^{c} ±0.06	3.04^{c} ±0.05	3.94^{a} ±0.11	3.31^{b} ±0.13
ABTS	7.40^{d} ±0.1	10.0^{bc} ±0.2	11.0^{a} ±0.2	10.2^{b} ±0.1	9.53^{c} ±0.3	10.9^{a} ±0.1	10.1^{b} ±0.1
ORAC	1160.6^{d} ±21	1128.8^{d} ±14	2303.3^{a} ±26	1618.2^{c} ±11	996.9^{e} ±4.1	2293.8^{a} ±9.8	1782.4^{b} ±4.8
Polyphenols	4.71^{f} ±0.1	7.82^{c} ±0.1	9.81^{a} ±0.4	6.78^{d} ±0.2	5.98^{e} ±0.2	9.37^{a} ±0.2	8.43^{b} ±0.2
Flavonoids	2.20^{e} ±0.04	3.70^{d} ±0.06	5.15^{b} ±0.02	3.62^{d} ±0.04	3.60^{d} ±0.04	5.56^{a} ±0.06	4.16^{c} ±0.05

Data are the mean of three independent experiments ± standard errors. *Different letters, in the same row, indicate significant differences at $p \leq 0.05$.

Table 4 Antioxidant activities (DPPH, µM Trolox Eq/g FW; ABTS, µM Trolox Eq/FW; and ORAC, µM Trolox Eq/100g FW) polyphenols (mg Tr/g FW) and flavonoids (mg rutin/g FW) in garlic bulbs grown for 3 months in soils differently treated: control CTR, soil without fertilizer; soil+sulphur+bentonite (SB); Sulphur+bentonite+orange residue (SBOr); Sulphur+bentonite+olive pomace (SBOp); Sulphur+bentonite+ municipal waste (SBMw); Sulphur+bentonite+orange residue+ municipal waste (SBOrMw); Sulphur+bentonite+olive pomace+ municipal waste (SBOpMw).

Garlic	**CTR**	**SB**	**SBOr**	**SBOp**	**SBMw**	**SBOrMw**	**SBOpMw**
DPPH	2.31^{e} ±0.01	3.44^{c} ±0.03	4.32^{a} ±0.05	3.25^{d} ±0.02	3.44^{c} ±0.02	3.77^{b} ±0.05	3.33^{cd} ±0.07
ABTS	9.4^{e} ±0.4	12.0^{c} ±0.3	13.1^{b} ±0.1	11.2^{d} ±0.2	11.5^{cd} ±±0.2	13.7^{a} ±±0.3	11.1^{d} ±0.1
ORAC	4301.6^{e}±5	5007.8^{b}±19	5468.3^{a}±21	5012.2^{b}±25	4890.9^{c}±17	5211.8^{ab}±18	4582.1^{d}±13
Polyphenols	6.91^{d}±0.5	8.82^{b}±±0.1	9.94^{a}±0.3	7.96^{c}±0.2	7.98^{c}±0.3	9.75^{a}±0.4	8.11^{c}±0.4
Flavonoids	4.20^{c}±0.3	5.60^{b}±0.3	6.41^{a}±0.2	5.95^{b}±±0.2	6.70^{a}±0.3	5.98^{b}±0.3	5.44^{b}±±0.3

Data are the mean of three independent experiments ± standard errors. *Different letters, in the same row, indicate significant differences at $p \leq 0.05$.

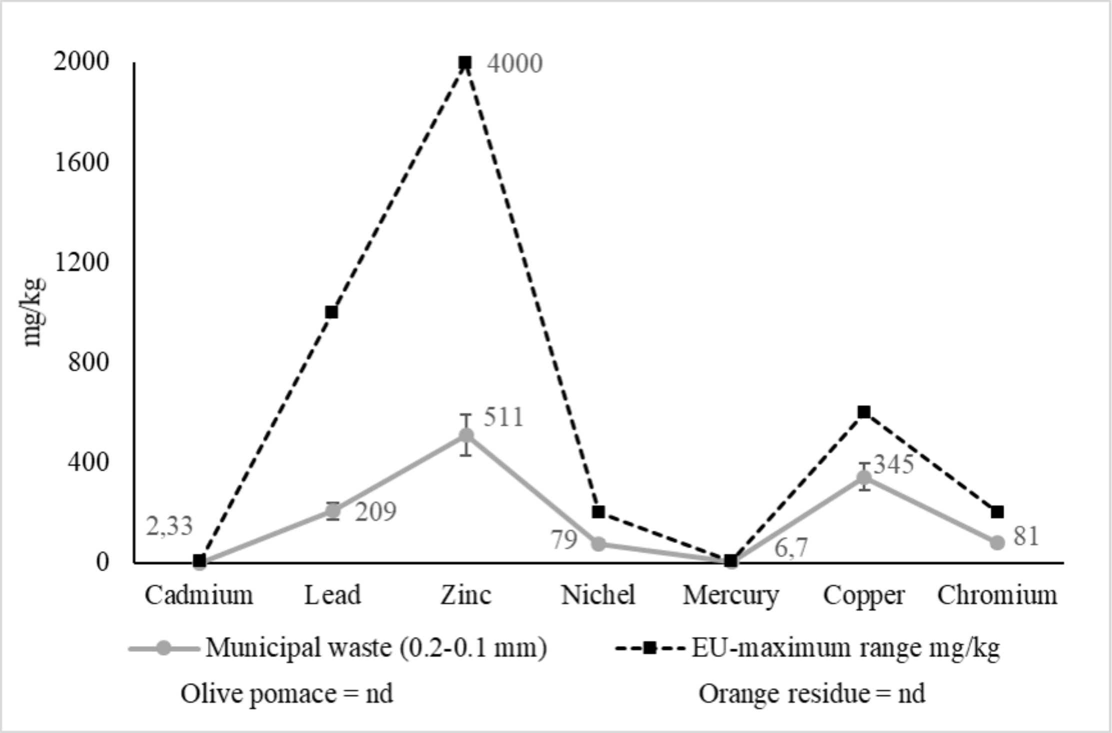

Figure 1 Content of cadmium, lead, zinc, nickel, mercury, copper and chromium (mg/kg) in Mw (0.2-0.1 mm), compared to the European maximum allowed limit. Or and Op didn’t contain heavy metals.

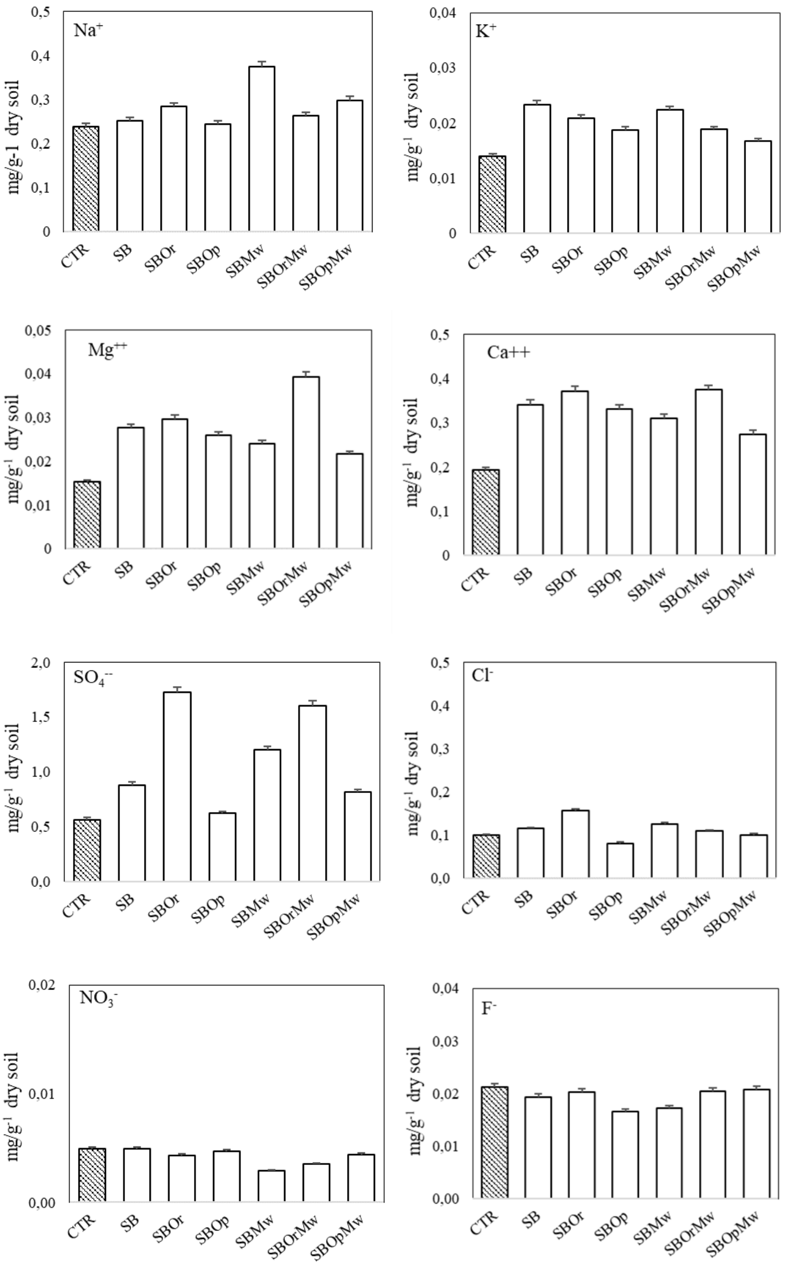

Figure 2 Cation and anion content in soil 3 months after the amendment with: CTR= Control, soil without fertilizer; soil+sulphur+bentonite (SB); Sulphur+bentonite+orange residue (SBOr); Sulphur+bentonite+olive pomace (SBOp); Sulphur+bentonite+municipal waste (SBMw); Sulphur+bentonite+orange residue+composted municipal waste (SBOrMw); Sulphur+bentonite+olive pomace+ dried municipal waste (SBOpMw). Data are the means of three independent experiments and bars represent the standard error of the parameters analysed.

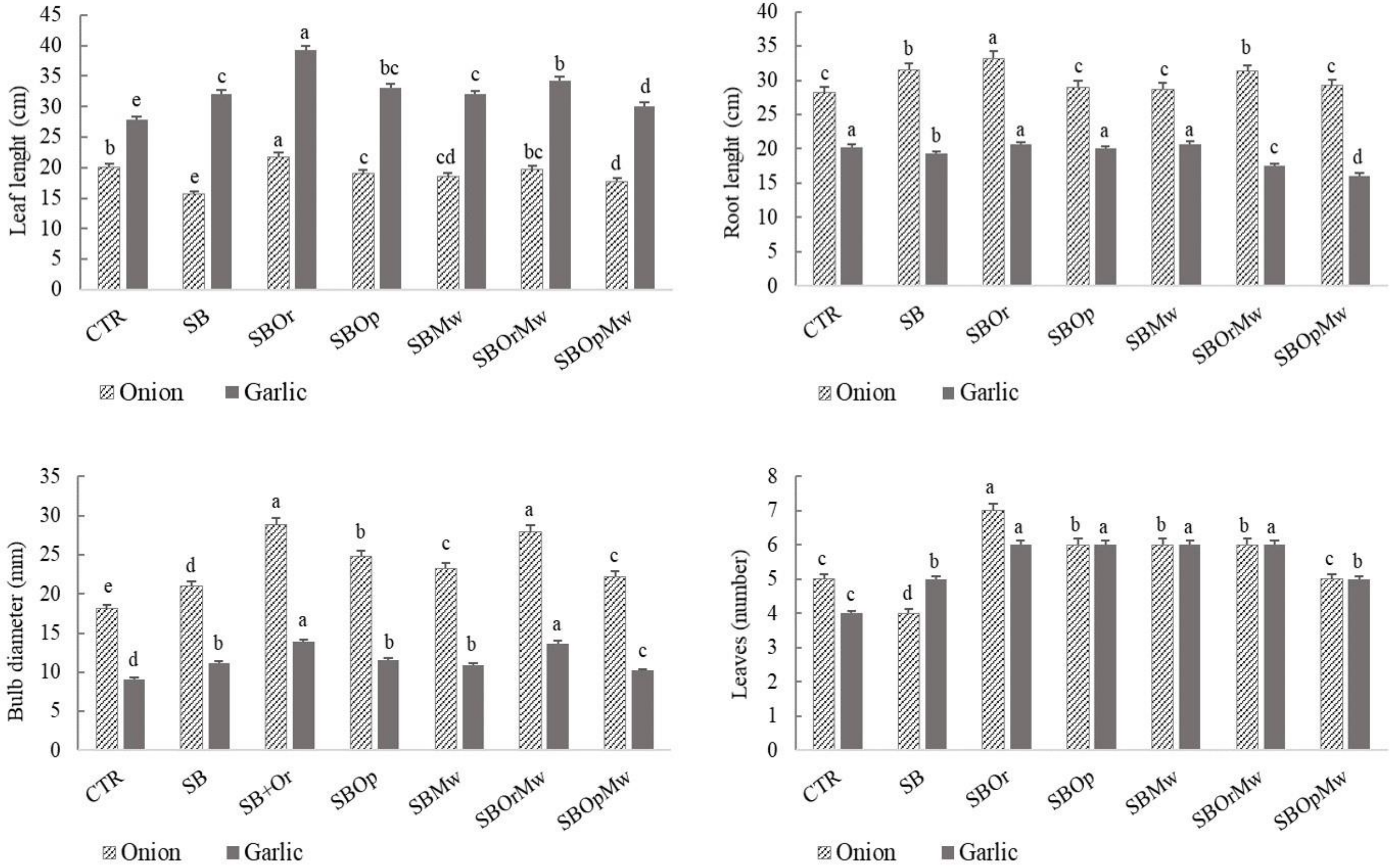

Figure 3 Growth parameters of onion and garlic grown for 3 months on soils differently treated. CTR= Control, soil without fertilizer; soil+sulphur+bentonite (SB); Sulphur+bentonite+orange residue (SBOr); Sulphur+bentonite+olive pomace (SBOp); Sulphur+bentonite+ municipal waste (SBMw); Sulphur+bentonite+orange residue+ municipal waste (SBOrMw); Sulphur+bentonite+olive pomace+ municipal waste (SBOpMw). Different letters, in the same group of bars, indicate significant differences at $p \leq 0.05$.