
Enhancing Soil Quality and Sustainability: A Comparative Study of Chemical, Organic and Waste-Based Fertilizers in Tomato-Grown Soils, with Emphasis on Carbon and Water Footprint

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Article

Enhancing Soil Quality and Sustainability: A Comparative Study of Chemical, Organic and Waste-Based Fertilizers in Tomato-Grown Soils, with Emphasis on Carbon and Water Footprint

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Abstract: This manuscript explores sustainable agriculture's role in mitigating environmental challenges and meeting the nutritional needs of a growing global population. The objective is to assess the impact of different fertilizers on two distinct tomato-growing soils, each characterized by unique chemical and biological properties. The study compares four fertilizer treatments: Additionally, it evaluates the environmental implications of these fertilizers, focusing on their Carbon and Water Footprints. The findings indicate significant improvements in soil quality with SBO fertilizer. Both soils experienced a shift towards a near-neutral pH, increased organic matter content, and heightened microbial biomass. Enzyme activities were notably superior in SBO-treated soils. Life Cycle Assessment (LCA) results demonstrate that the SBO-based system is the most sustainable, with the lowest Carbon footprint. NPK exerts the highest environmental impact. The Water Footprint analysis aligns with these results, showing that SBO requires the least water for tomato production. In summary, this study underscores the significance of sustainable fertilization practices in improving soil quality and reducing environmental footprints in agriculture, offering potential benefits for food production and environmental conservation.

Keywords: sustainability; soil quality; carbon footprint; water footprint

1. Introduction

Food systems currently account for approximately 33% of total human greenhouse gas (GHG) emissions, representing a significant and increasing share of global emissions. The emissions from agricultural activities and land use, estimated at around 12.0 GtCO₂ per year, were quantified by the Food and Agriculture Organization of the United Nations (FAO) and the Intergovernmental Panel on Climate Change (IPCC) [1]. These emissions are expected to rise further due to the growing demand for food. One of the primary contributors to GHG emissions is the production of synthetic nitrogen (N) fertilizers used in crop cultivation. The application of these fertilizers is widely recognized as a major factor in the release of nitrous oxide (N₂O) emissions from agricultural soils. N₂O is a potent greenhouse gas, with a global warming potential 265 times greater than carbon dioxide (CO₂) and methane (CH₄) [2].

Moreover, in the early 1900s, a process was developed to mass-produce a compound containing ammonia, which greatly increased crop yields while utilizing less land. Ammonia is now one of the most extensively produced chemicals globally and is used in large quantities as a highly effective fertilizer [3]. However, excessive, or improper application of nitrogen-based fertilizers can result in inefficient nitrogen uptake by plants, leading to an excess of nitrogen in the soil and subsequent N₂O emissions. According to the Intergovernmental Panel on Climate Change, the global use of synthetic nitrogen fertilizers has increased by 800% since the 1960s. The FAO estimates that this quantity is projected to increase by an additional 50% by the year 2050.

The European Union's commitment to achieving a climate-neutral economy by 2050 is outlined in the European Green Deal, which emphasizes the role of every sector in this transition [4].

Soil, being a major carbon accumulator, stores twice as much carbon as the atmosphere and three times as much as terrestrial biomass [5].

Therefore, implementing appropriate and sustainable fertilization practices that enhance carbon storage and reduce CH₄ and N₂O emissions is crucial for agriculture to mitigate climate change while maintaining soil productivity [6].

Numerous researchers have demonstrated that long-term balanced fertilization, including sulphur (S) along with other macro nutrients, can decrease N₂O emissions without compromising productivity, highlighting the significant role of elemental sulphur in improving and maintaining soil fertility.

Sulphur is recognized as a vital element for promoting optimal soil health and enhancing the availability of essential nutrients such as nitrogen, phosphorus, and potassium to plant roots. It deserves re-evaluation and consideration as a key component of soil fertility and productivity. Factors such as intensive cultivation practices, the use of sulphur-free fertilizers, and a decline in atmospheric sulphur depositions have contributed to sulphur deficiency in agricultural soils [7, 8]. Various studies have emphasized that sulphur deficiency leads to reduced nitrogen utilization from fertilizers and the production of crop proteins with significantly lower levels of sulphur-containing amino acids, particularly methionine, which greatly influences the nutritional value of plants [9, 10, 11].

Kulczycki et al., [12] provided evidence of the positive effects of elemental sulphur on plant yields and soil properties in crops such as mustard, wheat, rapeseed, and corn.

Further investigations revealed a negative association between the application of elemental sulphur and microbial activities in soils, leading to a significant decrease in microbial biomass carbon and enzyme activities [13].

This decline in microbial biomass and enzyme functions could potentially be attributed to a scarcity of carbon substrates, particularly in soils with low organic matter content. Conversely, numerous studies demonstrated the stimulating effects of organic amendments on soil microbial biomass and activities responsible for nutrient release [14, 15]. These findings have prompted researchers to explore the combination of sulphur and organic matter. In this regard, Tabak et al., [16] discovered that incorporating waste sulphur with organic materials facilitated the simultaneous enrichment of soil with readily available sulphur and organic matter. Moreover, Holatko et al., [17] conducted research showing that the utilization of elemental sulphur in conjunction with organic material, such as digestate, enhanced the availability of sulphur, resulting in increased yields and improved crop quality. Additionally, this combination was found to enhance the activity of soil microbes [18]. On the basis of the above considerations and of our previous works [19, 20, 21, 22] focused on the reuse of different kinds of biomasses for land restoration and crop improvement, this study aims to evaluate the effects of agro-industrial waste-based fertilizer on the quality, carbon footprint, and water footprint of two soils used for growing tomatoes, compared to commonly used chemical and organic fertilizers.

Tomato represents 13.5% of the world's vegetable production [23]. Several studies indicated that average GHG emissions for open-field tomato production were 0.2 e 2.0 kg CO₂-eq kg tomato⁻¹. [24] Hillier et al. [25] calculated the carbon footprint variation of different food crop production and found that applied N-fertilizer contributed most to variability. Lee et al., [26] performed an investigation to quantify the variability of GHG emissions due to soil properties and management and found that the field-scale variability of GHG emissions is controlled primarily by biochemical parameters rather than physical parameters. Results showed that differences in the variability for microbial activity as affected by tillage and irrigation develop different levels and combinations of field-scale controls on GHG emissions.

Aldaya and Hoekstra [27] analysed Italian industrial tomato production that had a WF equal to 114 m³ t⁻¹ (30% green, 50% blue and 17% grey). Comparing these results with Chapagain and Orr [28]

the blue component was in accordance with them, while green and grey were much higher in the Italian productive system, due to different weather conditions and fertilizers inputs.

The uniqueness of this research lies in the correlation between fertilizer efficiency, water usage, and greenhouse gas emissions, determined by assessing the Carbon footprint (CF) and Water footprint (WF). Simultaneous assessment of carbon and water footprint for agri-food products is desirable to provide better insights into key environmental issues than using either indicator in isolation [29]. These indicators measure the potential impact of a product throughout its entire life cycle in terms of greenhouse gas emissions and the consumption and degradation of water resources. The WF quantifies the volume of fresh water consumed or polluted (through evaporation or product incorporation) within a given time, while the CF indicates the Global Warming Potential over a defined period.

2. Materials and Methods

2.1. Agro-Industrial Waste-Based Fertilizer manufacturing

The fertilizer is composed by 85 % S pelletized with 10% bentonite clay (as carrier and inert support) and 5% of orange waste. After the pelletized phase, the mixtures have been introduced in a 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 pods. At the end they were obtained pads of diameter of 3/4 mm.

2.2. Soil treatment

Fertilizers were tested on Tomato plants, variety *Big Rio F1* and 30 cm diameter pots containing 9 kg of soil each were used. For the trial, 3 replications were made for each. The experimentation took place in a climatic cell to simulate the optimal environmental conditions for plant growth. The climatic conditions set in the climatic chamber for the germination phase were 25°C, 70% humidity and 800 Lux. For the vegetative development phase of the plants, climatic parameters were set following the climatic data from May until the end of August, considering that the vegetative cycle is 120 days for these varieties. Two fertilizations were carried out: one at the beginning of the crop cycle and one at complete flowering. The fertilizers used for each pos are:

- A) 1.2 g of synthetic fertilizer NPK (15-15-15) in which there are 0.18 g of N, 0.18 g of P₂O₅ and 0.18 g of K₂O.
- B) 13 g of organic fertilizer, in which there are 0.26 g of N, 0.26 g of P₂O₅ and 0.195 g of K₂O. In this fertilizer there are bovine, equine, sheep and poultry manure mixed with litter, calcium sulphate and olive pomace dust mixed at the total percentage of 50 % on composition.
- C) 1.4 g of fertilizer Sulfur-bentonite + orange waste.

2.3. Soil physical, chemical, and biochemical analysis before and after treatment

Potted soils before treatment were analyzed for physical and chemical properties. Soils were taken in the *Grecanica* area, specifically in the municipalities of Reggio di Calabria (RC) (soil 1) and Motta San Giovanni (RC) (soil 2). It was used a sandy-loam soil (soil 1) and a sandy-clay soil (soil 2) as stated by FAO soil classification system [30]. Soil texture has been detected by using the hydrometer method [31]. pH was measured in distilled water (soil/solution ratio 1:2.5) with a glass electrode. Electric conductivity (EC) was determined in distilled water by using 1:5 soil/water suspension mechanically shaken at 15 rpm for 1 h and then detected by Hanna instrument conductivity meter. CEC (Cation exchange capacity) was determined with Mehlich methodology [32]. Organic carbon was assessed with dichromate oxidation method [33] and transformed into organic matter multiplying by 1.72; Total nitrogen (TN) was measured with Kjeldahl method [34]. C/N was determined as a carbon/nitrogen ratio. Water soluble phenols were extracted in triplicate as reported by Kaminsky et al. [35]. Gallic acid was used as a standard and the concentration of water-soluble phenolic compounds was expressed as Gallic acid equivalents ($\mu\text{g GAE g}^{-1} \text{ d.s}$). Microbial

biomass carbon (MBC) was determined by using the chloroform fumigation-extraction procedure on fresh soil. [36] Fumigated and unfumigated soil sample extracts were used to detect soluble organic C using the methods of Walkley and Black [33]

Table 1. Physical and chemical properties of soils before fertilization.

	SOIL 1	SOIL 2
Skeleton (%)	45	21
Sandy %	65	50
Clay %	23	27
Loam %	12	23
Textural Class	Sandy-loam	Sandy-Clay-loam
Moisture %	18	32
S.S (%)	82	68
pH (H ₂ O)	8.5	8.3
pH (KCl)	7.8	7.3
EC (μS/cm)	107.3	302
CEC (cmol ⁽⁺⁾ kg ⁻¹)	21.57	27.71
TOC %	1.78	1.98
TN %	0.19	0.2
C/N	9.37	9.9
SOM %	3.07	3.41
WSP (μg GAE g ⁻¹ d.s)	56.1	85.62
MBC (μg C g ⁻¹ soil)	896	941

Dehydrogenase (DHA) activity was detected with iodinitrotetrazolium chloride according to von Mersi and Shinner method [37]. Catalase activity (CAT) was detected by the method of Kuush et al. [38] measuring the absorbance during the conversion of H₂O₂ to oxygen and water. The decrease in the absorbance was measured at 240 nm, utilizing the extinction coefficient of 39.4 M⁻¹ cm⁻¹. Fluorescein diacetate hydrolase (FDA) activity was determined according to the method of Adam and Duncan [39]. Beta glucosidase activity was tested as reported Valášková et al [40] with few modifications as reported in Muscolo et al. [19]. Soil (1 g fresh weight) was placed into a plastic tube and treated with 4 mL of modified universal buffer (MUB, pH 6). The reaction mixture contains 0.16 mL of 1.2 mM PNP substrate (p-nitrophenyl-β-D-glucoside) in 50 mM sodium acetate buffer (pH 5.0) and 0.04 mL of the sample. Reaction mixtures were incubated at 40 °C for 20–120 min. After incubation the reaction was stopped and the yellow color from the p-nitrophenol was developed by the addition of 0.1 mL of 0.5 M sodium carbonate, the p-nitrophenol absorbance was measured on a spectrophotometer at a wavelength of 400 nm and quantified by comparison with a standard curve. Protease activity was detected as reported by Sidari et al. [41]. Two ml of phosphate buffer (0.1 M, pH 7.1) and 0.5 ml of 0.03 M N-a-benzoyl-L arginin amide (BAA) have been added to 1 g of wet soil. The mixture was incubated at 37 °C for 1h and 30 min, then diluted to 10 ml with distilled water. The ammonium concentration was detected with an ammonium selective electrode (CRISON, micro-pH 2002). Urease activity was determined following the method of Kandeler and Gerber [42], with few modifications as reported in Sidari et al. [41]. Five grams of fresh soil were mixed with 2.5 ml of urea (80 mM) and 20 ml 0.1 M borate buffer at pH 10.0. After 2 h in an orbital shaker at 37 °C, 2.5 ml of urea were added to the control. add 30 ml of KCl (2 M) were instead added to both sample and control, shake for 30 min. One ml of the filtered solutions was mixed with 9 ml of distilled water, 5 ml of sodium/salicylate solution, and 2 ml of dichloroisocyanuric acid. Ammonium concentrations were determined at 690 nm by using a calibration curve. The results are reported as μg N-NH₄/g ds/3h. [41].

Table 2. Soil enzymatic activities before fertilization. Dehydrogenase (DHA, μg INTF g⁻¹ d.s h⁻¹). Catalase activity (CAT. O₂/3min/g d.s). Fluorescein diacetate hydrolase (FDA, μg fluorescein g⁻¹ d.s).

Urease (URE, N-NH₄/g ds/3h). β -glucosidase (β GLU, μ g para-nitrophenol (p-NP) g/h). Protease (PRO μ g Tyrosine *g d.s *2h).

	SOIL 1	SOIL 2
DHA	1.31	2.83
CAT	1.54	3.85
FDA	8.93	15.10
β -GLU	514	348
PRO	167	157
URE	312	289

2.4. Environmental impact: Carbon and Water footprint.

The study was carried in *Grecanica Area*, specifically in Motta San Giovanni, province of Reggio Calabria. The main features of the studied system were collected through visits to farm, direct interview with farmers using a specific collection sheet. The farm carried out the treatments by subdividing 4 experimental plots, carrying out the treatments described in section 2. The farm grew tomatoes of the *Big Rio F1* variety, with a planting density of 3 plants/m², with a sprinkler irrigation system. Pesticide, insecticide and acaricide treatments were the same for all experimental plots, as were soil tillage and harvesting. The only difference between the experimental plots was the fertilization. The farms inputs and outputs used in the analyzed systems are reported in table 3.

2.4.1. Carbon Footprint

The Life Cycle Assessment approach, according the ISO 14040:2006 [43]. was used to estimate environmental impacts with a focus on Global Warming (GWP 100a) for the Carbon Footprint.

LCA methodology contains four stages: goal and scope definition, life cycle inventory life cycle impact assessment and interpretation. [44] The system boundaries, as shown in **Figure 1**, starts with planting of tomato plants and ends with harvesting, that following the PCR 2010:07 "Arable and vegetative crops", corresponds to CORE processes, that are the farming phases: transports of plants, acceptance of plants, planting, harvest. In accordance with the PCR, [45] this study does not include: manufacturing of buildings and capital goods other than agricultural machinery, business travel of personnel, travel to and from work by personnel, research and development activities.

Furthermore, due to the lack of data, the analysis did not consider: input transport (fertilizers and pesticides) from the place of production to the field. All the inputs used in the agricultural phase (fertilizers, pesticides, fuels, various materials utilized during the harvesting) were considered. Primary data were collected in situ during the 2021-2022 agricultural year, from May to October, using a data collection sheet on a farm that carried out the experimentation in collaboration in the open field located in Motta San Giovanni (RC). All inputs are reported in table 3. Direct emissions from fuel and lubricant were taken from SimaPro's LCI database.

Table 3. Farm inputs and outputs used in the analyzed systems.

	CTR	A	B	C
Fertilizers (kg ha⁻¹)				
NPK		170		
Horse Manure			430	
Sulfur Bentonite + Orange waste				476
Chemicals (kg ha⁻¹)				
Primor 50	0.75	0.75	0.75	0.75
Daramun	4	4	4	4
Human labour (h ha ⁻¹)	45	46	48	46
Machinery (h ha ⁻¹)	5	10	12	10
Diesel (kg ha ⁻¹)	10	13	15	13
Water (m ³ ha ⁻¹)	750	750	750	750

Electricity (kWh kg ⁻¹)	0.13	0.14	0.15	0.15
Production (t ha ⁻¹)	47	57.5	58	58.4

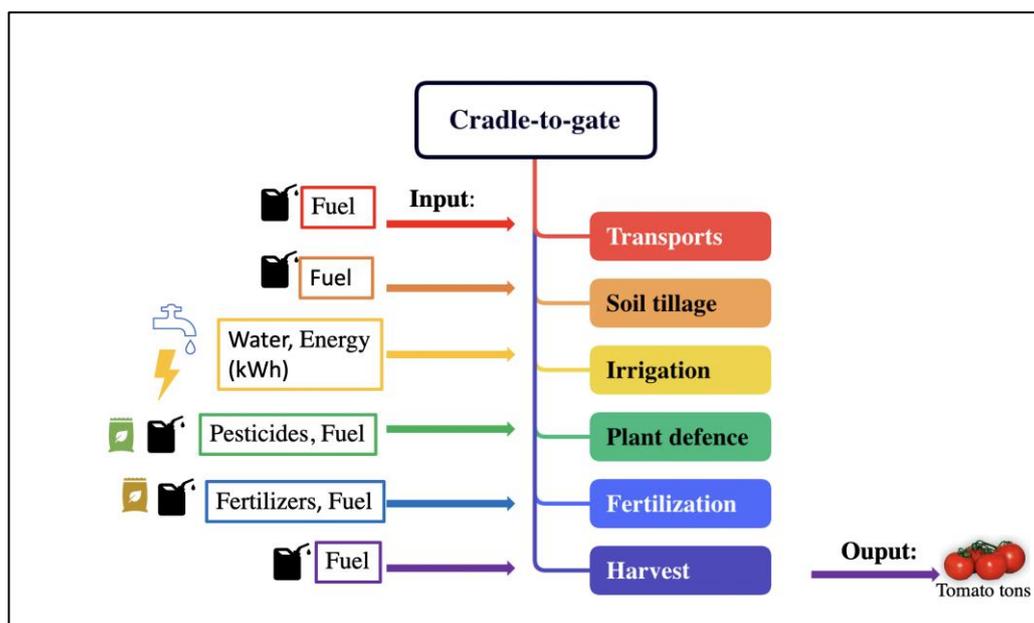


Figure 1. System boundaries of CORE process of tomato production.

The impact assessment was performed using SimaPro 9.02, with the problem oriented LCA method developed by the Institute of Environmental Sciences of the University of Leiden [46]. It was used the method CML-IA Baseline V3.06/ EU25 +3, 2000. The following impact categories were considered according to the selected method: abiotic depletion (AD); abiotic depletion (fossil fuels) (AD fossil fuels); global warming potential (GWP) or climate change; photochemical oxidation (PO); ozone layer depletion (ODP); human toxicity (HT); freshwater aquatic ecotoxicity (FWE); marine aquatic ecotoxicity (MAE); terrestrial ecotoxicity (TE); air acidification (AA) and eutrophication (EU).

2.4.2. Water Footprint

For this study WF was performed as established by Hoekstra et al. 2011 [47] in “The Water Footprint assessment manual” that defining the guidelines for the water footprint. The peculiarity of this methodology is the division of the water footprint into 3 components: blue, green, and grey. The total water footprint is given by the sum of them according to the formula below:

$$WF_{\text{total}} = WF_{\text{green}} + WF_{\text{blue}} + WF_{\text{grey}}.$$

Particularly, for the WFP blue and green is required the calculation of the crop water requirement (CWR, m³ ha⁻¹), following the formulas below:

$$WF_{\text{green}} = CWU_{\text{green}} / \text{crop yield}$$

$$WF_{\text{blue}} = CWU_{\text{blue}} / \text{crop yield}$$

The green and blue components of CWU (m³ ha⁻¹) were calculation by accumulation of daily evapotranspiration (ET mm/day) over the whole growing season:

$$CWU_{\text{green}} = 10 \times \sum_{d=1}^{d=\text{harvesting}} ET_{\text{green}}$$

$$CWU_{\text{blue}} = 10 \times \sum_{d=1}^{d=\text{harvesting}} ET_{\text{blue}}$$

ET_{green} represents green water evapotranspiration and ET_{blue} instead blue water evapotranspiration [47]. The summation is done over the period from the day of planting (day 1) to the day of harvest. The green and blue water evapotranspiration has been estimated by using the

CROPWAT model developed by the Food and Agriculture Organization of the United Nations [48] which is based on the method described by Allen et al. [49].

The CROPWAT model offers two different options to calculate evapotranspiration: the 'crop water requirement option' (assuming optimal conditions) and the 'irrigation schedule option' (including the possibility to specify actual irrigation supply in time [50] and for this study we use the first option. According to Pellegrini et al., [51]:

- ETgreen was calculated as the minimum of Crop Water Requirement (CWR, mm year⁻¹) and effective precipitation (Peff, mm year⁻¹).
- ETblue was estimated from Irrigation Requirement (IR) rates as the minimum between IR (m3 year⁻¹) and the irrigation volume (Ieff, m3 ha⁻¹ year⁻¹). [51].
- IR was calculated as a constant value for the analyzed systems according to the following equation: $IR = \max(0; CWR - Peff)$

In the CROPWAT 8.0 software, after entering the input data related to climate data of Reggio Calabria in the year 2021-2022, crop Kc, rainfall and soil characteristics we obtained the CWR needed to calculate the green and blue water footprint, as shown in Table 4.

Table 4. Crop water requirement obtained from CROPWAT 8.0.

Month	Decade	Stage	Kc	ETc	ETc	Eff rain	Irr. Req.
			coeff	mm/day	mm/dec	mm/dec	mm/dec
May	2	Init	0.6	2.24	13.4	8.4	6.4
May	3	Init	0.6	2.45	26.9	14.1	12.9
Jun	1	Init	0.6	2.7	27	14.6	12.3
Jun	2	Deve	0.64	3.12	31.2	14.3	16.9
Jun	3	Deve	0.78	3.76	37.6	13.6	24
Jul	1	Deve	0.93	4.31	43.1	12.4	30.8
Jul	2	Deve	1.07	4.92	49.2	11.4	37.8
Jul	3	Mid	1.18	5.9	64.9	12.8	52
Aug	1	Mid	1.18	6.61	66.1	14.9	51.2
Aug	2	Mid	1.18	7.14	71.4	16.3	55.2
Aug	3	Mid	1.18	6.65	73.1	15.6	57.6
Sep	1	Late	1.17	6.01	60.1	14.6	45.5
Sep	2	Late	1.07	5.1	51	14.1	36.9
Sep	3	Late	0.95	3.9	39	13.9	25
Oct	1	Late	0.85	2.83	17	7.3	10.9

Following Hoekstra et al. 2011 [47], the grey component of Water Footprint was calculated as:

$$WF \text{ product, grey} = [(\alpha \times AR) / (C_{\max} - C_{\text{nat}})] / Y$$

where:

- AR is the chemical application rate to the field per hectare (kg ha⁻¹);
- α is the leaching-run-off fraction;
- C_{\max} is the maximum acceptable concentration for the pollutant considered (kg m⁻³);
- C_{nat} is the natural concentration for the pollutant considered (kg m⁻³);
- Y is the crop yield (tons ha⁻¹).

For the grey component of WF only nitrogen fertilizers have been considered, according to Hoekstra et al., 2011, [47] because Nitrogen represents an important source of pollution in Europe as can be seen from the Nitrates Directive 1990. [51]. From Legislative Decree 152/2006 the acceptable limit value for nitrogen was found to be 15 mg/l and a leaching factor (α) 0.1 was considered for all cultivation systems.

3. Results

3.1. Effect of different fertilizers on soils quality

The chemical properties of soil 1, assessed six months after the treatments, exhibited significant variations. Notably, there were no significant changes in soil texture. However, in the soils treated with Sulfur Bentonite (SB), notable reductions were observed in both pH in water and KCl levels. Simultaneously, there was an increase in electrical conductivity (EC), suggesting the potential for an enhanced mineralization process with ion release. Additionally, the organic matter content, C/N ratio, cation exchange capacity (CEC), and microbial biomass all experienced increases. (Tab.5)

Conversely, in the soils treated with SB, the phenol content and catalase activity decreased compared to all other treatments, while other enzyme activities increased. A strong positive and significant correlation was observed between Microbial Biomass Carbon (MBC), organic matter, pH in KCl, C/N ratio, Water Soluble Phenol (WSP), FDA hydrolysis, Dehydrogenase (DHA) activity, Protease activity, and Urease activity. However, no correlation was found between MBC and beta-glucosidase, and MBC exhibited an inverse correlation with Catalase activity. (Tab.6)

Six months after the treatments, the chemical properties of soil 2 exhibited notable variations among the different treatment groups. However, in the soils treated with SB (Sulfur Bentonite), the reductions in both pH (measured in water) and KCl levels were particularly significant, surpassing the changes observed in soil 1. In contrast, Electrical Conductivity (EC), Cation Exchange Capacity (CEC), Organic Matter (OM), C/N ratio, Water-Soluble Phenols (WSP), and Microbial Biomass Carbon (MBC) all followed a similar increasing trend as seen in soil 1. (Tab.5)

When examining the biochemical data, akin to the findings in soil 1, the SB-treated soil in soil 2 exhibited elevated levels of DHA (Dehydrogenase), FDA (Fluorescein Diacetate), and Beta-glucosidase activities, surpassing the other treatments, as previously observed. However, in the case of Protease and Urease activities, they displayed increases compared to the control and NPK treatments, aligning with soil 1. (Tab. 6) Notably, they were found to be on par with the HM (Horse Manure) treatment, marking a departure from the results observed in soil 1.

Furthermore, Catalase activity once again registered its lowest values in the SB-treated soil, confirming the consistent pattern observed in soil 1.

Table 5. Physical and chemical properties of **soil 1** and **soil 2** six months after treatments with the different fertilizers: CTR= Control unfertilized soil; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue. Values are the mean of three replicates (n=12).

	CTR	A	B	C	CTR	A	B	C
	Soil 1				Soil 2			
Skeleton (%)	45 ^a	45 ^a	45 ^a	45 ^a	21 ^a	21 ^a	21 ^a	21 ^a
Sandy %	65 ^a	65 ^a	65 ^a	65 ^a	50 ^a	50 ^a	50 ^a	50 ^a
Clay %	23 ^a	23 ^a	23 ^a	23 ^a	27 ^a	27 ^a	27 ^a	27 ^a
Loam %	12 ^a	12 ^a	12 ^a	12 ^a	23 ^a	23 ^a	23 ^a	23 ^a
Textural Class	Sandy-loam	Sandy-loam	Sandy-loam	Sandy-loam	Sandy-Clay-loam	Sandy-Clay-loam	Sandy-Clay-loam	Sandy-Clay-loam
pH (H ₂ O)	8.3 ^a	8.76 ^a	8.0 ^b	7.82 ^b	8.2 ^a	8.43 ^a	7.89 ^{ab}	7.2 ^b
pH (KCl)	7.70 ^a	7.85 ^a	7.75 ^a	7.58 ^b	7.2 ^a	7.1 ^{ab}	7.4 ^a	6.98 ^b
EC (μS/cm)	106 ^b	123 ^{ab}	132 ^a	142 ^a	298 ^a	291 ^{ab}	267 ^b	326 ^a
CEC	20.65 ^b	22.87 ^b	23.65 ^{ab}	28.56 ^a	26.61 ^b	24.23 ^b	26.45 ^b	31.56 ^a
TOC %	1.67 ^b	1.45 ^b	2.02 ^a	2.03 ^a	1.51 ^b	1.25 ^b	1.98 ^a	2.12 ^a
TN %	0.14 ^a	0.17 ^a	0.16 ^a	0.12 ^{ab}	0.12 ^b	0.21 ^a	0.19 ^a	0.15 ^b
C/N	11.93 ^b	8.53 ^c	12.63 ^b	16.92 ^a	12.58 ^a	5.95 ^c	10.42 ^b	14.13 ^a
SOM %	2.88 ^b	2.50 ^b	3.48 ^a	3.50 ^a	2.60 ^{ab}	2.16 ^b	3.41 ^a	3.65 ^a
WSP (μg GAE*g ⁻¹ d.s)	55.68 ^a	55.23 ^a	52.45 ^b	51.53 ^b	82.67 ^b	79.81 ^b	91.76 ^a	97.23 ^a
MBC μg C g ⁻¹ soil	901 ^b	876 ^b	926 ^a	976 ^a	945 ^b	965 ^b	1023 ^{ab}	1198 ^a

Different letters in the same row indicate significant differences (Turkey's test $p \leq 0.05$).

Table 6. Enzymatic activities of Soil 1 and Soil 2 six months after treatments with the different fertilizers. CTR= Control. soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue. Dehydrogenase (DHA, $\mu\text{g INTF g}^{-1} \text{d.s h}^{-1}$), Catalase activity (CAT, $\text{O}_2/3\text{min/g d.s}$), Fluorescein diacetate hydrolase (FDA, $\mu\text{g fluorescein g}^{-1} \text{d.s}$), Urease (URE, $\text{N-NH}_4/\text{g ds/3h}$), β -glucosidase (βGLU , $\mu\text{g para-nitrophenol (p-NP) g/h}$), Protease (PRO $\mu\text{g Tyrosine *g d.s *2h}$). The data are the mean of three replicates (n=12).

	CTR	A	B	C	CTR	A	B	C
	Soil 1				Soil 2			
DHA	1.29 ^b	1.34 ^b	1.59 ^{ab}	2.01 ^a	2.69 ^b	3.1 ^{ab}	3.5 ^a	3.8 ^a
FDA	8.56 ^b	8.51 ^b	9.01 ^a	9.34 ^a	14.5 ^b	15.51 ^b	17.01 ^a	19.14 ^a
CAT	1.34 ^a	1.51 ^a	1.01 ^b	0.96 ^b	2.84 ^a	2.51 ^a	2.41 ^a	1.96 ^b
βGLU	510 ^a	546 ^a	555 ^a	557 ^a	355 ^b	365 ^b	372 ^{ab}	401 ^a
PRO	165 ^b	157 ^b	167 ^b	212 ^a	145 ^b	157 ^b	177 ^a	201 ^a
URE	298 ^b	312 ^{ab}	351 ^b	365 ^b	258 ^b	297 ^a	281 ^a	279 ^a

Different letters in the same row indicate significant differences (Turkey's test $p \leq 0.05$).

Table 7. Correlation matrix (Pearson (n)) of physical, chemical and biochemical properties of soil 1 six months after treatments.

from\ to	pH (H ₂ O)	pH (KCl)	EC	CEC	TOC %	TN %	C/N	SOM %	WSP	MBC	DHA	FDA	CAT	βGLU	PRO	URE
pH (H ₂ O)	1	0.853	-0.581	-0.638	-0.974	0.724	-0.938	-0.974	0.845	-0.931	-0.806	-0.902	0.972	-0.344	-0.768	-0.792
pH (KCl)	0.853	1	-0.398	-0.649	-0.718	0.978	-0.965	-0.718	0.618	-0.904	-0.770	-0.771	0.735	-0.075	-0.902	-0.560
EC	-0.581	-0.398	1	0.923	0.666	-0.278	0.600	0.666	-0.922	0.738	0.891	0.865	-0.719	0.944	0.700	0.952
CEC	-0.638	-0.649	0.923	1	0.635	-0.586	0.768	0.635	-0.871	0.858	0.970	0.897	-0.698	0.761	0.905	0.884
TOC %	-0.974	-0.718	0.666	0.635	1	-0.556	0.857	1.000	-0.902	0.884	0.795	0.911	-0.996	0.488	0.679	0.862
TN %	0.724	0.978	-0.278	-0.586	-0.556	1	-0.894	-0.556	0.468	-0.814	-0.682	-0.647	0.578	0.055	-0.875	-0.411
C/N	-0.938	-0.965	0.600	0.768	0.857	-0.894	1	0.857	-0.802	0.983	0.888	0.909	-0.877	0.311	0.932	0.755
SOM %	-0.974	-0.718	0.666	0.635	1.000	-0.556	0.857	1	-0.902	0.884	0.795	0.911	-0.996	0.488	0.679	0.862
WSP	0.845	0.618	-0.922	-0.871	-0.902	0.468	-0.802	-0.902	1	-0.894	-0.935	-0.977	0.932	-0.791	-0.771	-0.996
MBC	-0.931	-0.904	0.738	0.858	0.884	-0.814	0.983	0.884	-0.894	1	0.954	0.969	-0.913	0.482	0.943	0.860
DHA	-0.806	-0.770	0.891	0.970	0.795	-0.682	0.888	0.795	-0.935	0.954	1	0.974	-0.843	0.691	0.940	0.928
FDA	-0.902	-0.771	0.865	0.897	0.911	-0.647	0.909	0.911	-0.977	0.969	0.974	1	-0.942	0.672	0.880	0.960
CAT	0.972	0.735	-0.719	-0.698	-0.996	0.578	-0.877	-0.996	0.932	-0.913	-0.843	-0.942	1	-0.537	-0.728	-0.896
βGLU	-0.344	-0.075	0.944	0.761	0.488	0.055	0.311	0.488	-0.791	0.482	0.691	0.672	-0.537	1	0.428	0.843
PRO	-0.768	-0.902	0.700	0.905	0.679	-0.875	0.932	0.679	-0.771	0.943	0.940	0.880	-0.728	0.428	1	0.749
URE	-0.792	-0.560	0.952	0.884	0.862	-0.411	0.755	0.862	-0.996	0.860	0.928	0.960	-0.896	0.843	0.749	1

Table 8. Correlation matrix (Pearson (n)) of physical, chemical, and biochemical properties of soil 2 six months after treatment.

	pH (H ₂ O)	pH (KCl)	EC	CEC	TOC %	TN %	C/N	SOM %	WSP	MBC	DHA	FDA	CAT	βGLU	PRO	URE
pH (H ₂ O)	1	0,360	-	-0,966	-0,914	0,314	-	-0,914	-0,961	-0,964	-0,824	-0,922	0,849	-0,923	-0,919	0,127
pH (KCl)	0,360	1	-	-0,491	0,048	0,191	-	0,048	-0,111	-0,514	-0,207	-0,349	0,472	-0,524	-0,315	-0,168
EC	-0,583	-0,924	1	0,737	0,216	-	0,536	0,216	0,337	0,645	0,254	0,445	-0,495	0,606	0,415	-0,163

CEC	-0,966	-0,491	0,737	1	0,816	-0,511	0,853	0,816	0,867	0,918	0,666	0,814	-0,745	0,856	0,805	-0,286
TOC %	-0,914	0,048	0,216	0,816	1	-0,228	0,726	1,000	0,984	0,814	0,807	0,846	-0,718	0,769	0,858	-0,182
TN %	0,314	0,191	-0,506	-0,511	-0,228	1	-0,832	-0,228	-0,176	-0,131	0,278	0,075	-0,189	0,001	0,084	0,934
C/N	-0,765	-0,173	0,536	0,853	0,726	-0,832	1	0,726	0,693	0,592	0,285	0,458	-0,314	0,476	0,455	-0,735
SOM %	-0,914	0,048	0,216	0,816	1,000	-0,228	0,726	1	0,984	0,814	0,807	0,846	-0,718	0,769	0,858	-0,182
WSP	-0,961	-0,111	0,337	0,867	0,984	-0,176	0,693	0,984	1	0,903	0,878	0,925	-0,826	0,871	0,932	-0,073
MBC	-0,964	-0,514	0,645	0,918	0,814	-0,131	0,592	0,814	0,903	1	0,887	0,968	-0,948	0,991	0,961	0,109
DHA	-0,824	-0,207	0,254	0,666	0,807	0,278	0,285	0,807	0,878	0,887	1	0,974	-0,960	0,922	0,979	0,413
FDA	-0,922	-0,349	0,445	0,814	0,846	0,075	0,458	0,846	0,925	0,968	0,974	1	-0,978	0,980	0,999	0,261
CAT	0,849	0,472	-0,495	-0,745	-0,718	-0,189	-0,314	-0,718	-0,826	-0,948	-0,960	-0,978	1	-0,982	-0,973	-0,413
βGLU	-0,923	-0,524	0,606	0,856	0,769	0,001	0,476	0,769	0,871	0,991	0,922	0,980	-0,982	1	0,973	0,245
PRO	-0,919	-0,315	0,415	0,805	0,858	0,084	0,455	0,858	0,932	0,961	0,979	0,999	-0,973	0,973	1	0,259
URE	0,127	-0,168	-0,163	-0,286	-0,182	0,934	-0,735	-0,182	-0,073	0,109	0,413	0,261	-0,413	0,245	0,259	1

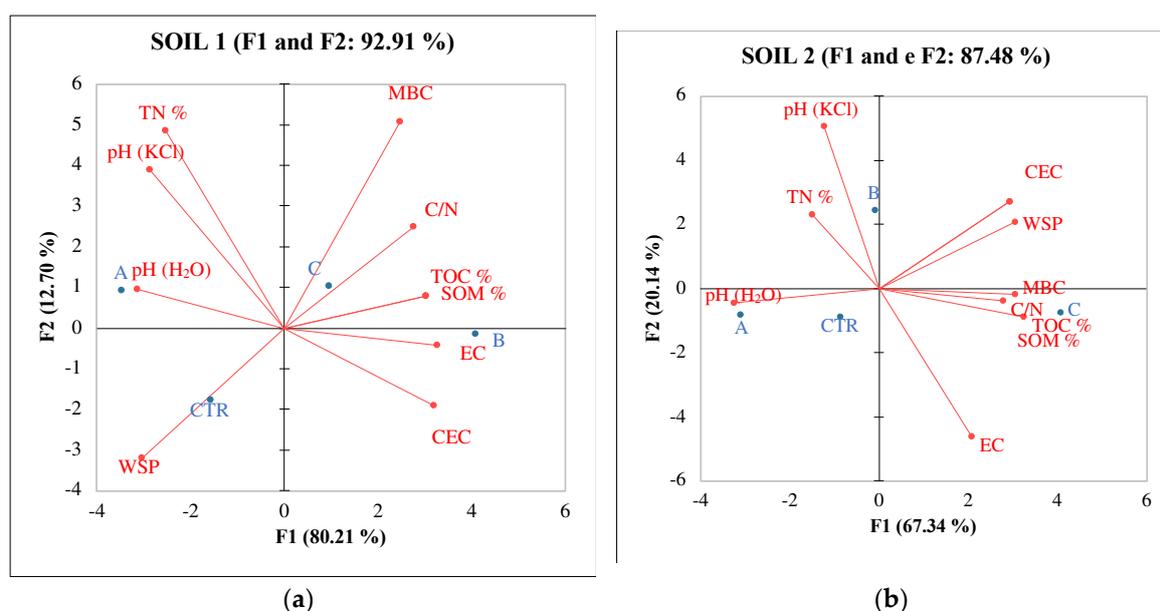


Figure 2. PCA of physical and chemical properties of soil 1 (a) and soil 2 (b) six months after treatments with the different fertilizers with CTR=Control, soil without fertilizer; A=nitrogen:phosphorous:potassium; B=horse manure; C=sulfur bentonite+orange residue.

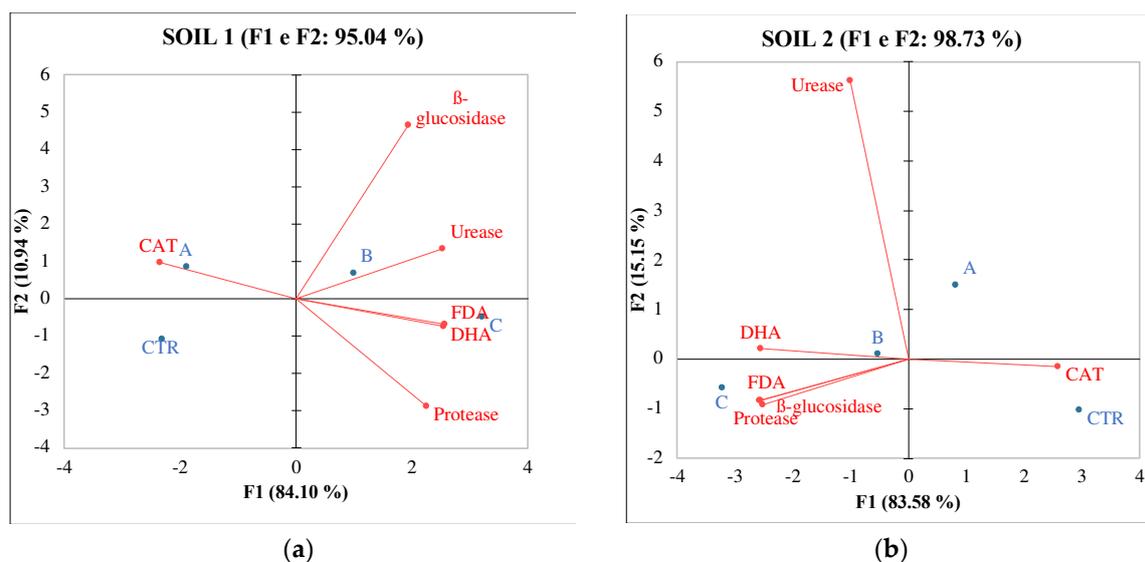


Figure 3. PCA of enzymatic activities of soil 1 (a) and soil 2 (b) six months after treatments with the different fertilizers CTR=Control, soil without fertilizer; A=nitrogen:phosphorous:potassium; B=horse manure; C=sulfur bentonite+orange residue .

3.2. Environmental impact

3.2.1. Carbon Footprint

The environmental analysis results, with a functional unit of 1 hectare and one ton of tomatoes considered for all impact categories for, can be found in Table 9 and Table 10. For Global Warming Potential (GWP 100a), Figure 4 reveals that among the four analyzed systems, System A has the highest impact at 26.04 kg CO₂ eq/ton. This is followed by System B at 23.21 kg CO₂ eq/ton and System CTR at 23.05 kg CO₂ eq/ton. Notably, System C, which utilized Sulphur fertilizer bentonite, stands out as the most sustainable with an impact value of 22.77 kg CO₂ eq/ton.

When breaking down by individual phases as seen in Figure 5, soil tillage emerges as the most impactful phase. This is followed by fertilization, registering an impact of 4.2 kg CO₂ eq/ton when applied. The organic fertilization using horse manure (System B) has an impact of 4.5 kg CO₂ eq/ton. Meanwhile, synthetic fertilization with NPK (System A) showcases the highest emissions among the systems, with values hitting 7.2 kg CO₂ eq/ton.

Table 9. Environmental impacts per hectare of analyzed system CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue.

Impact categories	Unit	CTR	A	B	C	
Abiotic depletion	kg Sb eq		0.01	0.02	0.01	0.01
Abiotic depletion (fossil fuels)	MJ	15055.69	17769.39	27543.07	15335.07	
Global warming (GWP100a)	kg CO ₂ eq	1083.46	1497.40	1346.10	1329.77	
Ozone layer depletion (ODP)	kg CFC-11 eq	0.00	0.00	0.00	0.00	
Human toxicity	kg 1.4-DB eq	494.38	650.97	530.92	508.03	
Fresh water aquatic ecotox.	kg 1.4-DB eq	523.45	634.97	540.73	534.12	
Marine aquatic ecotoxicity	kg 1.4-DB eq	629032.75	803512.81	678553.69	648589.00	
Terrestrial ecotoxicity	kg 1.4-DB eq	1.02	1.57	1.24	1.09	
Photochemical oxidation	kg C ₂ H ₄ eq	0.19	0.22	0.29	0.19	
Acidification	kg SO ₂ eq	7.36	15.95	9.74	8.77	
Eutrophication	kg PO ₄ --- eq	1.96	4.16	2.28	2.50	

Table 10. Environmental impacts for ton of tomato of analyzed system CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue.

Impact categories	Unit	CTR	A	B	C
Abiotic depletion	kg Sb eq		0.00	0.00	0.00
Abiotic depletion (fossil fuels)	MJ		320.33	309.03	474.88
Global warming (GWP100a)	kg CO2 eq		23.05	26.04	23.21
Ozone layer depletion (ODP)	kg CFC-11 eq		0.00	0.00	0.00
Human toxicity	kg 1.4-DB eq		10.52	11.32	9.15
Fresh water aquatic ecotox.	kg 1.4-DB eq		11.14	11.04	9.32
Marine aquatic ecotoxicity	kg 1.4-DB eq		13383.68	13974.14	11699.20
Terrestrial ecotoxicity	kg 1.4-DB eq		0.02	0.03	0.02
Photochemical oxidation	kg C2H4 eq		0.00	0.00	0.01
Acidification	kg SO2 eq		0.16	0.28	0.17
Eutrophication	kg PO4-- eq		0.04	0.07	0.04

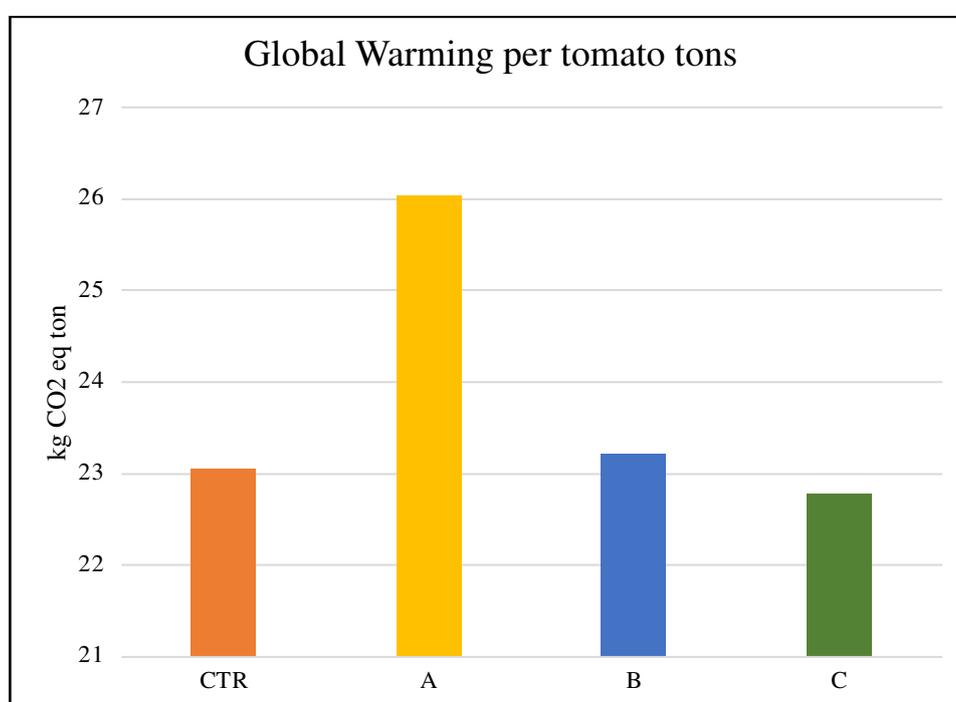


Figure 4. Global Warming of the entire life cycle in terms of kg of CO2 eq per tons of tomato. CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue.

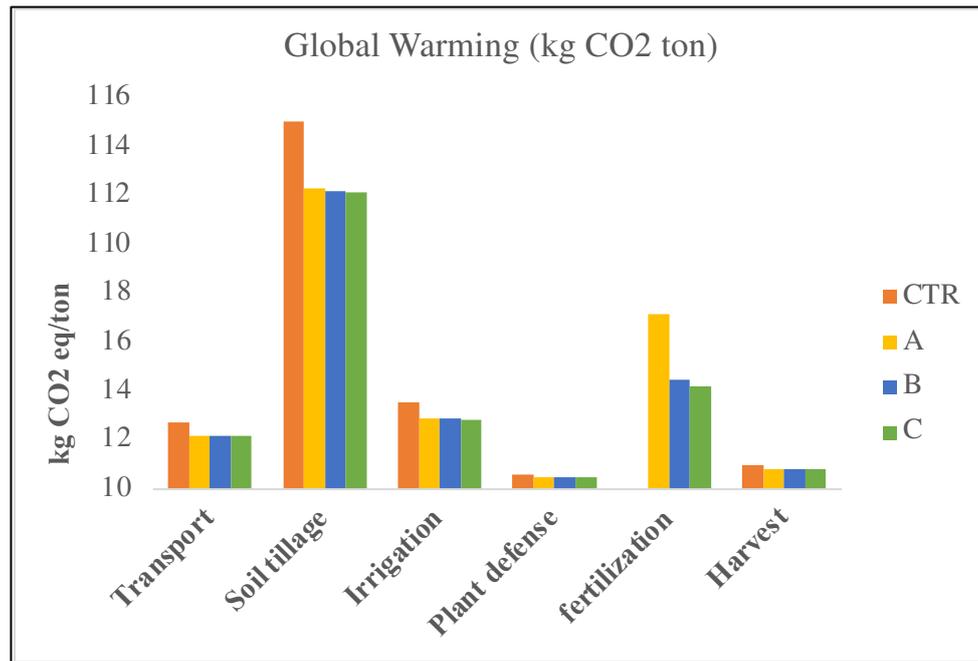


Figure 5. Global Warming per phase of the production process. Values per tons of tomato. CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue.

3.2.2. Water Footprint

The results of the water footprint (WF) are presented by breaking it down into its components (green, blue and grey) and then as total water consumption. Table 11 gives the data on yields, green evapotranspiration (Et green), blue evapo-transpiration (Et blue), direct and indirect fraction of water used for the different systems analysed, with the WF expressed in m³ per tonne of tomato yield. Since all systems are located in Reggio Calabria (RC) and share identical climatic conditions, green and blue evapotranspiration remains consistent across all systems and differences in productivity result mainly from different fertilisation approaches.

The system with the lowest water footprint is related to the use of sulphur-based bentonite fertilisers (A), which resulted in higher productivity. On the other hand, the control system (CTR) has the highest water footprint due to its lower production.

Table 11. WFP green and Blue of the analyzed systems. CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue.

Cultivation system	Yield (ton/ha)	Et green (m ³ /ha)	Et blue (m ³ /ha)	Direct and indirect fraction (m ³ /ha)	WFP green (m ³ /ton)	WFP blue (m ³ /ton)	WFP grey (m ³ /ton)
CTR	47.0	1983	1914	750.02	42.19	56.68	-
A	57.5	1983	1914	750.65	34.49	46.34	0.39
B	58.0	1983	1914	896.07	34.19	48.45	0.098
C	58.4	1983	1914	750.02	33.96	45.62	0.039

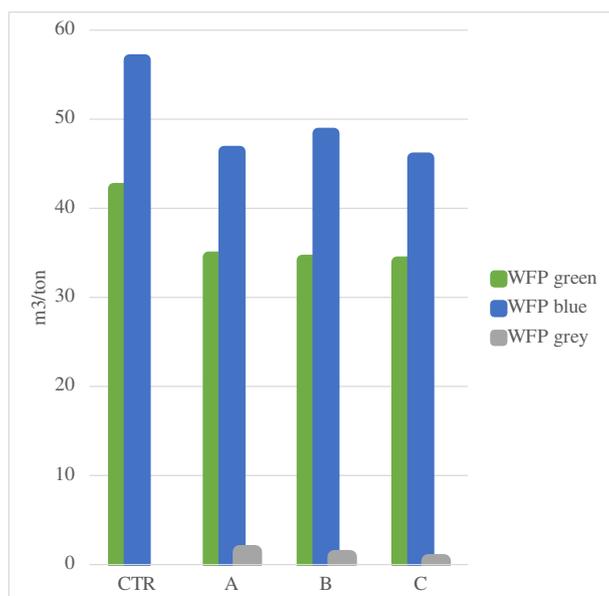


Figure 6. Total Water Footprint (m³/ton) broken down by its components (green, blue, grey) in the different cultivation systems. CTR= Control, soil without fertilizer; A= nitrogen:phosphorous:potassium; B= horse manure; C=sulfur bentonite + orange residue Values are expressed in m³ per tons of tomato.

4. Discussion

4.1. Effect of different fertilizers on soils quality

In both soil 1 and soil 2, the results indicate significant alterations in soil properties due to the application of Sulfur Bentonite (SB) fertilizer. These changes encompassed shifts in soil pH, reductions in KCl levels, increased electrical conductivity (EC), and notable enhancements in organic matter content, cation exchange capacity (CEC), and microbial biomass.

The observed decrease in phenol content and catalase activity in SB-treated soils compared to other treatments suggests that SB may have an impact on soil microorganisms and their metabolic activities. This is further supported by the positive correlations between Microbial Biomass Carbon (MBC), organic matter, pH in KCl, C/N ratio, Water Soluble Phenol (WSP), FDA hydrolysis, Dehydrogenase (DHA) activity, Protease activity, and Urease activity. These findings underscore the complex relationship between soil properties and microbial responses to fertilizer treatments.

In soil 2, similar trends were observed, with SB treatment leading to notable changes in soil properties and enzyme activities. However, the correlation pattern between MBC and other factors differed from that in soil 1, highlighting the influence of soil characteristics on the effects of added fertilizers.

Furthermore, the reduced Catalase activity in SB-treated soils suggests that SB does not induce stressful conditions in these soils, supporting its suitability for soil health and productivity. The complex relationship between catalase activity and microbial biomass is emphasized, influenced by factors such as soil pH, organic matter content, and temperature.

Principal Component Analysis (PCA) (Figure 2) results reveal multifaceted associations between SB fertilizer and soil properties, both chemical and biochemical. These associations differ between soil 1 and soil 2, possibly due to variations in initial soil conditions and microbial communities. Nonetheless, these findings provide valuable insights into the intricate interactions between fertilizers, soil characteristics, and microbial dynamics, essential for informed soil management practices and sustainable agriculture. Monitoring these parameters aids in assessing soil health, microbial activity, and nutrient cycling processes in differently treated soil ecosystems, contributing to environmental management.

4.2. Environmental impact

4.2.1. Carbon Footprint

The data offers a comparative understanding of the carbon footprint of various fertilization systems. System A's substantial impact underscores the environmental concerns surrounding synthetic fertilizers like NPK.

The significant reduction in carbon footprint when using Sulphur fertilizer bentonite (System C) indicates its potential as an environmentally friendly alternative. The distinctions between the systems and the high emissions linked to synthetic fertilization emphasize the necessity for transitioning towards more sustainable farming practices.

The evident emissions related to soil tillage and fertilization emphasize the environmental implications of these farming activities. Furthermore, the disparity in emissions between organic and synthetic fertilization methods warrants attention.

System B's horse manure-based organic fertilization produces fewer emissions than the synthetic NPK method in System A, highlighting the environmental advantages of certain organic fertilizers. However, the fact that System C's Sulphur fertilizer bentonite outperforms even the organic method suggests there are innovative solutions that can further reduce agriculture's carbon footprint.

4.2.2. Water Footprint

In the case of the cultivation system employing organic fertilizer (B), a higher impact is observed, mainly due to the direct fraction stemming from the water used in manure production. This indicates that organic fertilization may have implications for water use efficiency.

The results also reveal that the largest grey water footprint is associated with cultivation system A, which utilized synthetic NPK fertilizer. Conversely, due to the lower nitrogen content in sulfur bentonite fertilizer combined with orange residue, cultivation system C exhibits the lowest grey water footprint. This finding suggests that the choice of fertilizer can significantly impact the release of polluted water into the environment.

When examining the Total WFP, it becomes evident that the blue water footprint represents the largest percentage of water footprint when producing one ton of tomatoes for the various cultivation systems analysed, followed by the green footprint. This highlights the importance of considering both surface and groundwater consumption in assessing the overall water footprint of agricultural practices.

While the grey water footprint constitutes only 1% of the total water footprint, it is a crucial indicator as it signifies the quantity of polluted water released into the environment. This underscores the need for sustainable fertilization practices to minimize environmental pollution.

In summary, the results emphasize the environmental benefits of System C, where tomato plants were fertilized with sulfur-bentonite combined with orange waste, as a more environmentally friendly choice in terms of water footprint. These findings contribute to the understanding of how fertilizer choices can influence water use efficiency and environmental impact in agriculture.

5. Conclusions

In summary, this environmentally friendly fertilizer not only promotes ecological sustainability but also fosters economic growth rooted in renewable resources. This responsible utilization of resources aligns seamlessly with the core tenets of a green economy, which prioritize the preservation and efficient use of resources. By diminishing its environmental impact, this fertilizer contributes to the protection of ecosystems and biodiversity—a fundamental pillar of a green economy. This, in turn, safeguards the natural resources that form the bedrock of economic activities, particularly in agriculture. The sustainable production of this fertilizer can catalyze economic diversification by opening up new markets and opportunities within the agricultural sector. Such diversification can fortify local economies, making them more resilient.

Author Contributions: For research articles with several authors, a short paragraph specifying their individual contributions must be provided. The following statements should be used “Conceptualization, A.M (Adele Muscolo), A.M (Angela Maffia), GC; methodology, GC and A.M (Angela Maffia); software, A.M (Angela Maffia); validation, A.M (Adele Muscolo), G.C and C.M.; formal analysis, F.M.; investigation, M.R.; resources, F.V.; data curation, F.M.; writing—original draft preparation, A.M.; writing—review and editing, F.C.; visualization, A.M.; supervision, G.C.; project administration, A.M.(Adele Muscolo); funding acquisition, AM (Adele Muscolo); All authors have read and agreed to the published version of the manuscript.” Please turn to the CRediT taxonomy for the term explanation. Authorship must be limited to those who have contributed substantially to the work reported.

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