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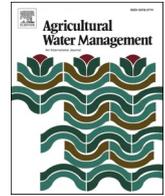
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Impacts of wastewater irrigation on Mediterranean soil and food: A three-year case study in Italy

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ABSTRACT

Water supply for irrigation is a limiting factor for agriculture in Mediterranean countries. A strategy for increasing water availability proposes to use low quality water for irrigation to avoid irrigation with precious and high cost potable water. A drawback about using waste water is the potential heavy metal accumulation in soil and foodstuff. In this study, primary or secondary treated municipal wastewater went through supplementary cleaning steps with a low-cost device and was used to irrigate tomatoes and potatoes in an experimental field in Italy. We monitored heavy metals in soil, food and processed food during three years. We measured no accumulation neither in soil, nor in food after this period. Variations of metal concentrations in soil and food were mainly due to common farm practices such as fertilisation and pesticide applications. This study demonstrates that irrigation with wastewater is achievable with a low-cost treatment device that can be used elsewhere to decrease pressure on water resources.

1. Introduction

Water availability for irrigation is the principal limiting factor for the increase of agricultural production in the Mediterranean basin, indispensable to meet the general increase in food demand (EU Common Agricultural Policy 2023–2027). A possible strategy is based on the reuse of wastewater for agricultural irrigation to reduce the amount of water that needs to be extracted from conventional water sources (Al-Hazmi et al., 2023).

The use of water in agriculture is more and more frequently conflicting with environmental uses and with the achievement of WFD objectives in water-scarce areas. To address water reuse over Europe, providing a supportive and coherent legal framework, in 2022, the European commission published Guidelines to support the application of Regulation 2020/741 on minimum requirements for water reuse (2022/C 298/01), setting rules for reuse water in agriculture. The treated wastewater (TWW) use for agricultural production, which is already a common and global practice (Paranychianakis et al., 2015; Sato et al., 2013), is expected to extend.

Nevertheless, some countries and stakeholders are reluctant to embrace the practice of water reuse since the long-term use of wastewater for irrigation could impact the soil composition especially

concerning inorganic compounds (Cajuste et al., 2002; Singh, 2021). Some significant heavy metal accumulation was observed in soils irrigated over decades with the raw sewage (Aslam et al., 2023; Dère et al., 2006).

Indeed, from a European perspective, considering efforts to improve water quality, the expected TWW quality has to be defined locally, identifying specific risk and their mitigation actions, avoiding any impact of the irrigation on soil. In addition, as the European consumer's interest in high quality food is increasing, the impact of TWW on produced irrigated food has to be limited and current regulations to be complied with, notably the Regulation (EU) 2023/915 on maximum levels for certain contaminants in food.

The key of using TWW for irrigation appears to further treat water at appropriate costs in line with the requirements of Directive 91/271/EEC governing the Urban Waste Water Treatment Directive in order to meet the new minimum quality parameters and become suitable for use in agriculture. This is not always economically feasible in a Waste Water Treatment Plant (WWTP). Treating wastewater up to drinking water quality for irrigation is a solution that will eliminate or greatly reduce such risks but will enhance treatment costs to a level not affordable by the farmers.

A balance between the two extremes, low-cost raw WW and WW

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treated to drinking-water level, has to be found to propose to farmers an economically viable and safe set of technological options for the reuse of wastewater (Huerta et al., 2002). To achieve this balance, innovative technology (Battilani et al., 2009, 2010) and water management options (Plauborg et al., 2010) were tested to lower the risks linked to the use of low quality water (EU FP6 SAFIR project-<https://cordis.europa.eu/project/id/23168/reporting>).

During the project, an innovative low-cost system to clean municipal wastewater for irrigation was tested in an experimental field site in Italy. Specifications of this technology are reported in Battilani et al. (2009, 2010). The aim of this paper is to evaluate the impact of irrigation water coming from this treatment system on soil quality. The evaluation focused not only on soil quality, but also on accumulation of inorganic compound in tomatoes, and potatoes irrigated with treated wastewater, as well as in puree and potatoes flakes produced from these crops at the experimental field site in Italy.

The inorganic compounds studied were As, B, Cd, Cu, Cr, Fe, Ni, Pb, Zn. The main specificities of the present study are:

- Specific low-cost treatment and polishing steps after primary and secondary treatment were designed and tested.
- The irrigation water quality was such as can be realistically expected after appropriate and economically viable treatment.
- Relevant quality parameters were monitored throughout the system, from raw wastewater to processed food (potato flakes and tomato puree) over all intermediate steps (treated effluents, irrigation water, soil, and raw crops).
- Cropping was realised at plot scale simulating real farm practices. The experimental plots were large enough to be cropped like normal farm plots, applying normal fertilisation amounts and irrigation quantities.
- The monitoring was performed during over 3 irrigation seasons (2006, 2007, 2008) to assess the inter-annual variations and cumulative effects of inorganic compounds contents in soil and crops.

2. Materials and methods

2.1. Site characteristics

The field was located nearby the village of Mezzolara di Budrio (Bologna) in the plain of the Po valley (44°34 N, 11°32 E). The field was part of a commercial farm close to the small waste water treatment plant serving the village (Fig. 1). The field was drained with coated drain laid down every 13 m at a depth of 0.8–1.0 m. The drains discharged in a manifold located at the north end of the field. The soil has a high content of silt and clay (in average 48 % and 47 % respectively) and some fine

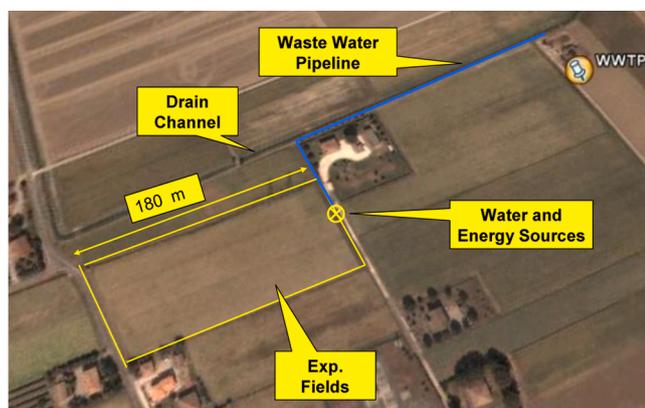


Fig. 1. Aerial view of the experimental fields with the situation of the waste water treatment plant (WWTP), water adduction and drainage water evacuation channel.

sand. A shallow water table was present most of the time at a depth ranging from -0.60 m to -1.50 m, only exceptionally lower (1.8 m).

2.2. Water treatment system

Vegetables were irrigated with primary (PTWW) or secondary (STWW) treated municipal wastewater (Table 1) going through supplementary cleaning steps specifically developed for use on the project field sites (one step for PTWW and two for STWW). Tap water (TW) was used as a reference for the irrigation of a selection of plots.

The treatment devices providing complementary treatment of input wastewater are:

a compact pressurised Membrane Bioreactor prototype (MBR), developed by Grundfos, Denmark, directly treating PTWW. The MBR treated PTWW by active sludge in a pressurised environment. The active sludge volume stored inside each MBR reactor is of about 300 L.

Untreated wastewater was screened to remove the unwanted material, normally found in the sewage water. The raw wastewater was then pre-treated over a 100–200 mm mesh screen, automatically cleaned, to achieve the treatment.

The polishing step for STWW consisted in a commercial gravel filter system (coarse sand of 1.2–1.7 grain diameter, Netafim, Israel). It differs from agricultural filters due to its short water retention time and discontinuous flow, although the filtration process is similar.

Both treatments were coupled with drip irrigation with buried drip lines.

The wastewater monitoring during the first year revealed that both PTWW and STWW did not contain inorganic compounds in sufficiently high amounts to properly test the treatment devices and that the input function was highly variable.

In consequence, during years 2 and 3 both the inlet PTWW and STWW were spiked with selected heavy metals and metalloids. The spike was realised by adding a flow of an inorganic compound stock solution to the input water. This flow was proportional to the input water flow, so that the following constant target concentrations entering the treatment systems (MBR/sand filter) were to be reached: $20 \mu\text{g L}^{-1}$ in As, $5 \mu\text{g L}^{-1}$ in Cd, $100 \mu\text{g L}^{-1}$ in Cr, $200 \mu\text{g L}^{-1}$ in Cu, $100 \mu\text{g L}^{-1}$ in Pb. The stock solution was prepared by adding solid salts containing the inorganic target compounds to tap water and was then added to the input water flow using a MixRite injection pump assuring proportional metering. HM-spiked input waters were then treated by MBR or commercial sand filter.

A detailed technical description of the device used and an analysis of the performance of the treatment procedures is provided in previous works (Battilani et al., 2010; 2009).

Table 1

Concentration of selected inorganic elements in PTWW and STWW before treatment.

	PTWW		STWW	
	Avg	Std Dev	Avg	Std Dev
As [$\mu\text{g L}^{-1}$]	23.8	11	3.6	6.3
B [$\mu\text{g L}^{-1}$]	319.3	203	354	538.4
Cd [$\mu\text{g L}^{-1}$]	5.9	2.8	1.3	2
Cr [$\mu\text{g L}^{-1}$]	113.4	53.9	12.7	32.1
Cu [$\mu\text{g L}^{-1}$]	257.4	116.1	46.2	78.8
Fe [$\mu\text{g L}^{-1}$]	0.5	0.4	0.2	0.5
Ni [$\mu\text{g L}^{-1}$]	5.7	2.5	5.5	9.8
Pb [$\mu\text{g L}^{-1}$]	111	53.6	12.2	33.8
Zn [$\mu\text{g L}^{-1}$]	217.2	327	135.9	207.4
Ca [mg L^{-1}]	87.3	26.4	97.7	23.3
K [mg L^{-1}]	10	4.2	7.2	5.6
Mg [mg L^{-1}]	20.1	6.1	21.3	4.2
Na [mg L^{-1}]	105.2	72.7	65.6	52.9

2.3. Experimental plan

The experimental field extended over a length of 180 m and was divided into plot test combinations of water treatment (TW, PTWW, STWW) with irrigation types (mini-sprinkler, sub-surface drip irrigation) and irrigation strategies (full irrigation and partial root drying (PRD) in year 1, regulated deficit of irrigation (RDI) and PRD in year 2 and 3) in triplicate. The regulated deficit of irrigation plots received around 20 % less water than fully irrigated one (Shahnazari et al., 2007).

On half of the plots, the rotation was potatoes/tomatoes/potatoes, on the other half the rotation was tomatoes/potatoes/tomatoes. The sampled plots were fully irrigated in year 1; RDI, now the prevailing irrigation practice in Italy, was applied in year 2 and year 3 thus approaching real farming conditions, compared to the experimental PRD. Each plot was irrigated for three years with the same water source (TW, PTWW or STWW). Water quality shifts due to technological modifications, the details on the irrigation management and the analysis of the performance of the treatment procedures are documented in Jensen et al. (2010).

2.4. Potatoes lay down and irrigation

Potatoes of the Agata variety were cropped. Seed tubers were planted at a depth of 8 cm below field level and ridged with 10–15 cm of soil. The distance between ridges was 75 cm. When plants emerged and had reached a height of 3–5 cm, they were ridged a second time with 7–8 cm of soil. Netafim Uniram® self-compensating drip lines (diameter 17 mm) were placed on top of the ridges and secured with clamps. The drip lines were buried during the second ridging. The distance between emitters was 30 cm and they were positioned in the middle between two plants. Their discharge rate was 1.6 L h⁻¹.

Fertiliser was applied via fertigation during the plant growth cycle. Neither organic nor starter fertilisers were used. The amount of nutrients was determined from measurement of the N, P and K status of the soil in spring and by means of nutrient balances. Irrigation and fertigation were managed by means of the FertOrgaNic DSS model, specifically developed for potato and calibrated and validated in the area.

Chemical treatments, fungicide and pesticide treatments and weeds control were applied to avoid pests and fungi and weeds infestation.

2.5. Tomatoes lay down and Irrigation

Tomatoes of the mid-late harvested Perfect Peel variety were cropped. Tomatoes were mechanically planted in double rows with a spacing between plants of 40 cm, the double rows were spaced of 150 cm (density: 33,000 plants/ha). Netafim Uniram® self-compensating drip lines (diameter 17 mm) were positioned in the middle of the double line. The distance between emitters was 40 cm and these were positioned exactly in the middle between two plants. Their discharge rate was 1.6 L/h. The drip lines were buried on top of the ridges at an approximate depth of 10 cm. Irrigation and fertigation were managed by means of the Fertirrigere V3.2 model, specifically developed for processing tomato and calibrated and validated in the area. Moreover, a starter fertilisation with a small amount of Phosphor and nitrogen was placed below or beside (50 mm to 150 mm) the plant at planting. The insects and fungi were controlled by chemical treatments. Weeds were mainly controlled by herbicides but some mechanical interventions were also realised.

2.6. Heavy metal water sampling and analyses

In order to achieve a representative measurement of the concentration of inorganic compounds in the irrigation waters, a Proportional Like Device (PLD) was set up to collect integrated samples. Samples were collected either after the MBR or after the sand filter, upstream of the

fertigation injector. Samples were collected as well in the TW irrigation system as controls. The PLD was regulated to catch water only during the irrigation time (few hours a day). Before each integration period, a tank was fixed to the PLD. A sufficient quantity of concentrated suprapure® HNO₃ was added to the tank beforehand to reach pH 2 at the end of the sampling duration. Acidification stabilised metals for one month and more and gave access to the total input of contaminants to the plot (suspended and dissolved). Samples were filtered at 0.45 µm, stored in PE bottles. For the water samples, concentrations were measured in the BRGM laboratories. Major element (Ca, K, Mg, Na) concentrations were measured by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectroscopy, NF EN ISO 11885). The accuracy for all element concentrations is 5 %, except Na and K with 10 %. Other elements (As, B, Cd, Cu, Cr, Fe, Ni, Pb, Zn) were measured by ICP-AES (NF EN ISO 11885) or ICP-MS (Inductively Coupled Plasma Mass Spectrometry, NF EN ISO 9963–1) with variable accuracy ranging from 5 % to 15 %.

2.7. Heavy metal soil sampling and analyses

In the drip-irrigated plots, the pattern of potential contamination by low quality irrigation water is not evenly distributed but follows the drip irrigation lines. Samples were not taken randomly but within the soil volume of potential influence around each of the drip emitters. A total of at least 5 subsamples, for a total mass of 1000 g, were taken to form a composite sample. Taking a rather low number of subsamples allowed to minimise soil disturbance and to vary the sampling points from one campaign to the other (e.g. sampling around every second drip emitter...). The sampling depth integrated over the whole root zone and samples were mixed and homogenised before analysis. Total concentrations were determined after dissolution. Elemental soil analyses were performed after alkaline fusion with Na₂O₂ at 450 °C during 1 h and subsequent dissolution with HCl. For the soil samples, concentrations were measured in the BRGM laboratories. Major element (Ca, K, Mg, Na) concentrations were measured by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectroscopy, NF EN ISO 11885). The accuracy for all element concentrations is 5 %, except Na and K with 10 %. Other elements (As, B, Cd, Cu, Cr, Fe, Ni, Pb, Zn) were measured by ICP-AES (NF EN ISO 11885) or ICP-MS (Inductively Coupled Plasma Mass Spectrometry, NF EN ISO 9963–1) with variable accuracy ranging from 5 % to 15 %.

2.8. Food analyses

The applied protocols are based on the CODEX Alimentarius Commission and FAO/WHO Food Standards Programmes, norms ISO 10381–1, ISO 10381–2 and internal protocols (SSICA /PPC-12/1998-PR).

Homogeneous fresh samples (potato puree and tomato puree) were prepared according to the following extraction protocol: samples were washed with ultrapure grade water, sample peels were removed by means of an automatic potato peeler (SSICA protocol). Then, to obtain a homogeneous puree for analysis, each representative peeled sample was cut and homogenised under vacuum with a high-speed cutter system for 2 min. On the homogenised potato samples, cations (Ca, K, Na) and metals/metalloid (As, B, Cd, Cr, Fe, Ni, Pb, Sn, Zn, Cu), concentrations were determined in the SSICA laboratories by ICP-OES and ICP-MS (X Series 2, ThermoScientific) analysis respectively following standard ISO 11885 after microwave acid digestion. N concentration in potato puree samples was measured by the Kjeldhal Method. The same analytical protocols were used for fresh matter and processed food.

2.9. Statistical tests

Statistical tests were realised with MS EXCEL. Similitude between samples was verified with a two-ways ANalysis Of the VAriance (ANOVA). ANOVA was used to decide whether sets of samples were

different or not by comparing their averages. The ANOVA computes the value of a variable F; the larger the value of F, the more likely it is that at least one of the set of samples has an average different from the others. If F falls below a fixed threshold, it can be concluded that all the sets of samples have equal averages. A 0.05 level of probability was used to calculate the critical value of F.

3. Results and discussion

In the following, we compare and interpret the inorganic compounds analysed in soil, crops, and process foods at the end of the last irrigation seasons for each crops (tomatoes then potatoes).

Tomatoes plots: soil, crop and processed food compositions.

3.1. Evolution of the soil composition with tomato cropping

Despite the differences of composition of the irrigation water (Table 2), mean soil contents for all individual inorganic elements are similar for the three types of irrigated plots at the end of the Year 3 (29th August Year 3) (Table 3).

A sample collected in the TW irrigated plot and concentrated at 501 mg of Pb/kg of soil was discarded as outlier. This punctually important increase exclusively for Pb contents can be explained by lead shotgun pellets and its degradation products (Cao et al., 2003a,b), and by the common practice of hare hunting in the area.

Observing the variation over three years (19th June Year 1–29th August Year 3), only small differences appear between the three sets of plots which have been cultivated twice with tomatoes and once with potatoes.

No differences have been measured between soil irrigated with STWW and tap water. The only significant difference is that the K₂O content decreases stronger in the plots irrigated with PTWW. The F value for each element is below the critical value expect for K₂O, only for PTWW (Table 3).

K is an indispensable element for plants, and is associated with movement of water, nutrients, and carbohydrates in the plant tissue. A decrease of K in soil is probably the sign of the tomato plant uptake since this compound is a vital element for plant growth and metabolism (Hasanuzzaman et al., 2018; Liu et al., 2021). Nitrate uptake has been shown to stimulate net K⁺ uptake in various crop species, suggesting that the NO₃⁻ ion serves as a mobile accompanying anion during K⁺ uptake and/or transport (Fang et al., 2020). PTWW water was the richest in NO₃, it could therefore be expected an increase of the K uptake by root (Zhang et al., 2010). For all other trace and major inorganic compounds, variations in the three set of plots are similar and relatively small. The observed differences of irrigation water quality do not show any impact on the soil chemistry.

The absence of apparent impacts of irrigation water quality does not

imply that soils concentrations did not evolve in three years. For instance, the content in Ni increases by 34 % in plots irrigated with TW and by 30 % in plots irrigated by PTWW.

This parallel evolution of soil chemistry (Fig. 2), independently of irrigation water quality, suggests that the observed variations are driven by factors more significant than irrigation water quality variability. The 2-ways ANOVA analyses confirm this hypothesis, for the majority of the compound analyses there is a clear difference between the year and not between the type of water use for irrigation (Table 3). The fertilisation of plots and the applications of pesticides could explain these variations to a certain extent, particularly when considering the impurities contained in those farming products (Atafar et al., 2010; Defarge et al., 2018).

3.2. Evolution of tomatoes composition

The third year, the tomatoes harvested are very similar whatever their irrigation modes, the main differences are the content in P content between TW irrigated tomatoes and PTWW-irrigated tomatoes as well as Cr content between TW-irrigated tomatoes and STWW-irrigated tomatoes. The F value for these elements is above the critical value.

It seems that there is no clear connection between these differences of concentration and the quality of the irrigation water. PTWW and STWW are more concentrated in Cr than TW but only tomatoes irrigated with STWW have different concentration in this compound. The concentrations of the third year cannot be clearly linked to irrigation water quality (Table 4).

Even if irrigation water quality does not seem to have an impact, tomatoes harvested in Year 3 are significantly different from the tomatoes harvested in Year 1 (Fig. 3). The variations are very important (up to 600 % for Cd) and similar for TW, PTWW and STWW irrigated tomatoes except for some compound like Ni. A 2-ways ANOVA analyses indicate clearly that, like for soil, most of the analysed compound vary from one year of cropping to the other, but not between the types of water treatment (Table 4).

The year-to-year variations in inorganic compound are sometimes important but similar for TW, PTWW and STWW irrigated tomatoes. For instance, the variation in nutrient concentrations in tomatoes can be attributed to several factors, including differences in ripeness and cell turgor during fruit development, which particularly affect potassium (K) uptake. Lead (Pb) in the soil can influence calcium (Ca) levels in tomato roots, and high concentrations of cationic nutrients like Ca²⁺ and magnesium (Mg²⁺) can inhibit K uptake by competing for binding sites on root surfaces.

Our results differ from previously published data. It has been shown that high concentrations of certain soils can affect vegetation. However, some inorganic compounds can inhibit the adsorption of other inorganic compounds (Antosiewicz, 2005). For instance, increase of B is expected to reduce the Zn up taken by the plant (Leece, 1978).

Table 2

Concentration of selected inorganic elements in TW, PTWW and STWW used for tomatoes irrigation ("<": below limit of quantification).

	year 1			year 2			year 3		
	TW	PTWW	STWW	TW	PTWW	STWW	TW	PTWW	STWW
As[μg/l]	< 1.00	2.40	1.50	< 2.00	7.00	< 2.00	< 0.10	2.53	9.95
B[μg/l]	139.00	168.00	167.00	113.50	146.50	116.50	96.67	1031.33	2005.50
Cd[μg/l]	< 0.50	< 0.50	< 0.50	1.00	1.55	0.50	< 0.10	1.10	3.45
Cr[μg/l]	< 0.50	< 0.50	< 0.50	1.00	2.80	1.00	0.17	7.90	40.75
Cu[μg/l]	9.00	4.00	4.00	3.45	13.75	15.50	1.50	35.53	131.50
Fe[mg/l]	0.04	0.02	0.03	0.02	0.05	0.04	0.03	0.08	0.08
Ni[μg/l]	4.00	3.70	2.00	1.30	5.30	2.70	0.53	8.67	5.75
Pb[μg/l]	< 1.00	< 1.00	1.60	< 1.00	1.05	< 1.00	0.30	0.97	42.85
Zn[μg/l]	164.00	57.00	40.00	58.50	85.50	157.50	6.53	88.53	213.95
Ca[mg/l]	111.10	72.70	52.40	93.30	59.25	70.10			
K[mg/l]	2.40	7.30	8.00	2.00	5.45	3.70			
Mg[mg/l]	24.50	17.10	13.20	19.10	13.05	14.65			
Na[mg/l]	29.00	65.60	56.60	24.15	45.90	36.15			

Table 3
2-ways ANOVA analyse - Element concentration in soil before the first planting and after harvest of the third year.

	Soil before cultivation in year 1		Soil after cultivation in year 3		ANOVA	
	irrigated with TW (n = 6)	irrigated with PTWW (n = 6)	irrigated with TW (n = 6)	irrigated with PTWW (n = 6)	Effect of Year	Effect of Treatment
As [mg/kg]	<LQ	<LQ	35.83 ± 4.36	39 ± 4		
B [mg/kg]	76 ± 8	79 ± 9	82 ± 16	78 ± 18	0.17 (0.68)	0 (1)
Cd [mg/kg]	<LQ	<LQ ¹	<LQ	<LQ		
Cr [mg/kg]	119 ± 11	115 ± 11	141 ± 10	139 ± 12	27.52 (<0.05)	0.39 (0.54)
Cu [mg/kg]	40.17 ± 6.49	41.67 ± 3.44	46.33 ± 4.27	44.17 ± 1.33	6.08 (<0.05)	0.04 (0.85)
Fe2O3 [%]	4.8 ± 0.41	4.75 ± 0.45	5.08 ± 0.55	5 ± 0.6	1.65 (0.21)	0.1 (0.75)
Ni [mg/kg]	51.67 ± 5.72	51.50 ± 6.02	69.17 ± 5.53	66.83 ± 5.91	48.09 (<0.05)	0.28 (0.6)
Pb [mg/kg]	11.33 ± 1.51	10.50 ± 0.84	25.50 ± 5.61	24.80 ± 3.27	73.6 (<0.05)	0.19 (0.67)
Zn [mg/kg]	110.17 ± 9.39	110.83 ± 7.86	118.17 ± 8.13	116.83 ± 8.7	4.03 (0.06)	0.01 (0.92)
CaO [%]	9.33 ± 0.23	8.93 ± 0.45	9.55 ± 0.33	9.47 ± 0.60	4.67 (<0.05)	1.94 (0.18)
K2O [%]	2.73 ± 0.2	3.62 ± 0.19	2.88 ± 0.24	2.82 ± 0.18	15.12 (<0.05)	23.87 (<0.05)
MgO [%]	2.37 ± 0.25	2.38 ± 0.22	2.53 ± 0.29	2.48 ± 0.29	1.52 (0.23)	0.02 (0.88)
Na [%]	0.66 ± 0.06	0.68 ± 0.09				
P2O5 [mg/kg]	2188 ± 246	2217 ± 259	2944 ± 85	2832 ± 243	58.25 (<0.05)	0.22 (0.65)
	irrigated with TW (n = 6)	irrigated with STWW (n = 6)	irrigated with TW (n = 6)	irrigated with STWW (n = 6)		
As [mg/kg]	<LQ ¹	<LQ ¹	35.83 ± 4.36	39.67 ± 3.61		
B [mg/kg]	76 ± 8	78 ± 8	82 ± 16	83 ± 13	1.27 (0.27)	0.11 (0.75)
Cd [mg/kg]	<LQ ¹	<LQ ¹	<LQ ¹	<LQ ¹		
Cr [mg/kg]	119 ± 11	118 ± 9	141 ± 10	144 ± 9	35.71 (<0.05)	0.1 (0.76)
Cu [mg/kg]	40.17 ± 6.49	39.5 ± 5.17	46.33 ± 4.27	45.67 ± 1.75	10.12 (<0.05)	0.12 (0.73)
Fe2O3 [%]	4.8 ± 0.41	4.85 ± 0.38	5.08 ± 0.55	5.13 ± 0.36	2.59 (0.12)	0.08 (0.78)
Ni [mg/kg]	51.67 ± 5.72	53.50 ± 2.35	69.17 ± 5.53	69.67 ± 6.47	61.49 (<0.05)	0.3 (0.59)
Pb [mg/kg]	11.33 ± 1.51	10.83 ± 1.6	25.50 ± 5.61	24.20 ± 2.17	75.03 (<0.05)	0.41 (0.53)
Zn [mg/kg]	110.17 ± 9.39	113.5 ± 7.18	118.17 ± 8.13	121.00 ± 6.32	5.86 (<0.05)	0.93 (0.35)
CaO [%]	9.33 ± 0.23	9.27 ± 0.41	9.55 ± 0.33	9.32 ± 0.33	0.98 (0.33)	1.24 (0.28)
K2O [%]	2.73 ± 0.2	2.9 ± 0.19	2.88 ± 0.24	2.88 ± 0.1	0.75 (0.4)	1.17 (0.29)
MgO [%]	2.37 ± 0.25	2.4 ± 0.22	2.53 ± 0.29	2.55 ± 0.21	2.5 (0.13)	0.06 (0.81)
Na [%]	0.66 ± 0.06	0.68 ± 0.06				
P2O5 [mg/kg]	2188 ± 246	2171 ± 421	2944 ± 85	2831 ± 179	43.45 (<0.05)	0.36 (0.55)

("<LQ": all values below limit of quantification)

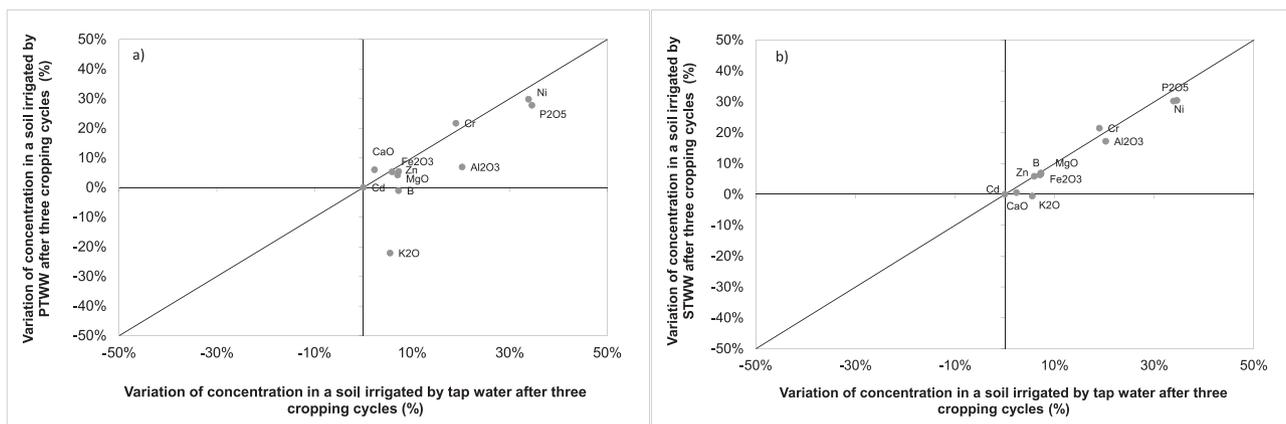


Fig. 2. Comparison of the concentration variation of the inorganic compounds in the soil, under tomato cropping, after three cropping cycles. Comparison between tap water irrigation and a) PTWW, and b) STWW.

That could imply that in multi element-containing soils, like those of this experiment, the plant concentration of a given compound would not be as high as in a soil polluted with only one single compound (Balba et al., 1991). That may imply that compound- and crop-specific experimental thresholds have to be increased before seeing any visible effect on the edible part (Zhu et al., 2007).

In our specific case, thanks to the treatment system used, these thresholds may not have been reached, while they may have been in other studies (Aslam et al., 2023; Singh et al., 2010)

3.3. Tomatoes puree food composition

Despite the differences of composition of the irrigation water, the tomato purees produced with tomatoes harvested at the end of the Year

3 (29th August Year 3) are similar (Table 5). The F values for all elements are below the critical value.

The variations of some compounds during food processing are very important. Comparing Year 3, the B content in raw tomatoes decreases by more than -50 % during processing of the tomatoes irrigated with TW. A boron decrease is expected due to its role as structural cross link in pectin polysaccharides as the galacturonic acid (Shi et al., 2017) that are gradually encrusted during the fruit development in the tomato skin to link the cuticle to the epidermis. Fruit peeling during the process will cause significant losses of B. On the other hand, concentrations in some elements increase during processing, for instance K and N.

From the conclusions on raw food, we can expect no link between irrigation water quality and processed food trace element contents.

No clear trend is discernible that might link modifications of

Table 4
2-ways ANOVA analyse - Element concentration in tomato fruits for each harvest depending of the water treatment.

	Tomatoes cultivated in year 1		Tomatoes cultivated in year 3		ANOVA	
	irrigated with TW	irrigated with PTWW	irrigated with TW	irrigated with PTWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.05 ¹	0.03 ¹	<LQ	<LQ		
B [mg/kg]	-	-	6.06 ± 0.21	4.47 ± 1.45		
Cd [mg/kg]	0.02 ¹	<LQ	0.08 ± 0.01	0.06 ± 0.01		
Cr [mg/kg]	0.04 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.07 ± 0.02	20.39 (<0.05)	0.02 (0.89)
Cu [mg/kg]	0.84 ± 0.40	1.00 ± 0.09	1.12 ± 0.13	1.21 ± 0.13	7.59 (<0.05)	1.82 (0.19)
Fe [mg/kg]	3.22 ± 0.87	3.15 ± 1.1	1.94 ± 0.62	2.71 ± 0.26	7.28 (<0.05)	1.25 (0.28)
Ni [mg/kg]	0.04 ± 0.02	0.04 ± 0.02	0.2 ± 0.12	0.12 ± 0.04	22.8 (<0.05)	2.03 (0.17)
Pb [mg/kg]	0.01 ¹	<LQ	0.26 ± 0.08	0.17 ± 0.02		
Sn [mg/kg]	0.11 ²	0.13 ¹	0.76 ± 0.1	0.66 ± 0.12		
Zn [mg/kg]	0.88 ± 0.16	1.09 ± 0.31	2.5 ± 0.9	2.54 ± 0.2	58.56 (<0.05)	0.39 (0.54)
K [mg/100 g]	195 ± 10	202 ± 10	375 ± 58.03	319.33 ± 12.88	142.74 (<0.05)	3.73 (0.07)
Na [mg/100 g]	-	-	4.22 ± 1.54	3.24 ± 1.01		
N [mg/100 g]	126 ± 24	125 ± 7	157.67 ± 17.8	173.67 ± 10.98	36.61 (<0.05)	1.19 (0.29)
P [mg/100 g]	52.9 ± 4.2	49.3 ± 2.2	58.52 ± 3.74	36.58 ± 4.02	5.74 (<0.05)	73.61 (<0.05)
	irrigated with TW	irrigated with STWW	irrigated with TW	irrigated with STWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.05 ¹	0.05 ¹	< 0.10	< 0.10		
B [mg/kg]	-	-	6.06 ± 0.21	0.00		
Cd [mg/kg]	0.02 ¹	0.01 ²	0.08 ± 0.01	0.08 ± 0.03		
Cr [mg/kg]	0.04 ± 0.01	0.05 ± 0.03	0.07 ± 0	0.11 ± 0.03	21.5 (<0.05)	5.78 (<0.05)
Cu [mg/kg]	0.84 ± 0.40	0.93 ± 0.14	1.12 ± 0.02	1.26 ± 0.16	10.28 (<0.05)	1.36 (0.26)
Fe [mg/kg]	3.22 ± 0.87	3.2 ± 1.66	1.94 ± 0.39	2.53 ± 0.85	4.91 (<0.05)	0.43 (0.52)
Ni [mg/kg]	0.04 ± 0.02	0.04 ± 0.01	0.2 ± 0.01	0.24 ± 0.16	19.26 (<0.05)	0.2 (0.66)
Pb [mg/kg]	0.01 ¹	0.01 ¹	0.26 ± 0.01	0.16 ± 0.07		
Sn [mg/kg]	0.11 ²	0.20 ²	0.76 ± 0.01	0.72 ± 0.17		
Zn [mg/kg]	0.88 ± 0.16	1.00 ± 0.2	2.5 ± 0.81	1.89 ± 0.52	33.12 (<0.05)	1.26 (0.28)
K [mg/100 g]	195 ± 10	207 ± 7	375 ± 58.03	354.33 ± 39.49	126.91 (<0.05)	0.07 (0.79)
Na [mg/100 g]	-	-	4.22 ± 1.54	3.33 ± 1.1		
N [mg/100 g]	126 ± 24	122 ± 20	157.67 ± 17.8	181 ± 16.15	31.32 (<0.05)	1.46 (0.24)
P [mg/100 g]	52.9 ± 4.2	51.2 ± 3.3	58.52 ± 3.74	47.69 ± 15.45	0.09 (0.76)	3.31 (0.08)

("<LQ": all values below limit of quantification; 1 = only one value above LQ; 2 = only one values above LQ)

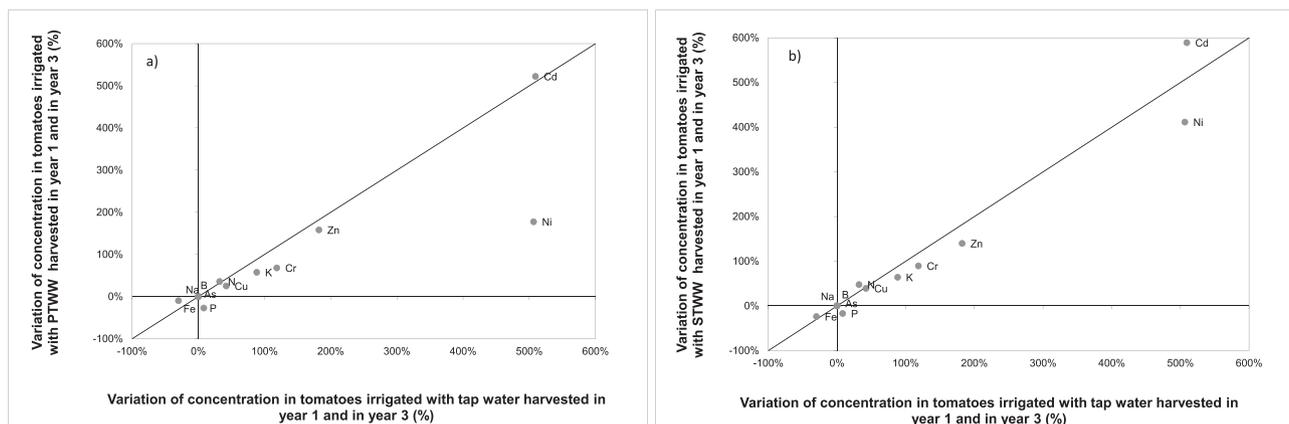


Fig. 3. Comparison of the concentration variation of the inorganic compounds in tomatoes after three cropping cycles. Comparison between tap water irrigation and a) PTWW, and b) STWW.

concentrations to the food processing even if an underlying parallel evolution of concentration is measurable (except for B).

Potatoes plots: soil, crop and processed food compositions.

3.4. Soils composition with potatoes cropping

In the case of the plots cultivated twice with potatoes and once with tomatoes, the difference over three years (4th April Year 1–14 th July Year 3) between the three types of soils irrigated with different water types are not significant (Table 7) despite the differences between water concentrations (Table 6). The F value for each element is below the critical value for the soil of the last year except for Pb.

At the end of the third irrigation season, no clear influence of the water quality on soils can be stated.

Important variations of the soil composition were measured for almost all the compounds.

The content of almost all the compound evolved similarly in the three set of soils. The most important exception between the soil sets concerns Pb (+3 % in STWW irrigated plots, -10 % in the two others sets of plots). The observed chemical differences between irrigation waters do not explain the soil composition variation over the total duration of the experiment. Except for Pb, the three sets of soil evolved in a similar way (Table 6 and Fig. 4).

Over the three years, variations in water quality were not uniform whereas the variations of soil inorganic compounds contents are parallel for the two sets of plots as it was the case for tomatoes plots. A 2-ways ANOVA analyses shows that differences are significant between the year of cropping and not between the types of water treatment for most

Table 5
2-ways ANOVA analyse - Element concentration in tomato puree for each harvest depending of the water treatment.

	tomatoes puree processed in year 1		tomatoes puree processed in year 3		ANOVA	
	irrigated with TW	irrigated with PTWW	irrigated with TW	irrigated with PTWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.05 ± 0.02	0.05 ²	< 0.10	< 0.10		
B [mg/kg]	-	-	2.67 ± 0.74	2.67 ± 0.41		
Cd [mg/kg]	0.01 ± 0 ³	0.01 ± 0 ³	0.07 ± 0.01	0.09 ± 0.04	60.56 (<0.05)	0.47 (0.5)
Cr [mg/kg]	0.13 ± 0.09	0.08 ± 0.03	0.17 ± 0.09	0.12 ± 0.04	1.02 (0.33)	1.82 (0.2)
Cu [mg/kg]	1.62 ± 0.23	1.66 ± 0.36	2.08 ± 0.18	2.07 ± 0.42	11.79 (<0.05)	0.01 (0.91)
Fe [mg/kg]	4.73 ± 0.68	4.93 ± 1.78	3.48 ± 0.47	3.63 ± 0.76	8.88 (<0.05)	0.16 (0.7)
Ni [mg/kg]	0.07 ± 0.03	0.09 ± 0.03	0.27 ± 0.07	0.34 ± 0.11	68.16 (<0.05)	2.23 (0.15)
Pb [mg/kg]	0.18 ± 0.15	0.07 ± 0.05	0.28 ± 0.05	0.24 ± 0.08	12.6 (<0.05)	3.74 (0.07)
Sn [mg/kg]	0.06 ± 0.03	0.1 ± 0.05	0.96 ± 0.15	0.95 ± 0.07	312.38 (<0.05)	0.01 (0.91)
Zn [mg/kg]	2.33 ± 0.26	2.47 ± 0.44	3.90 ± 0.30	3.91 ± 0.31	16.1 (<0.05)	0.11 (0.75)
K [mg/100 g]	224.4 ± 7.1	223.2 ± 18.6	485.3 ± 18.6	480.3 ± 29.1	1014.02 (<0.05)	0.14 (0.71)
Na [mg/100 g]	5.7 ± 1.4	6.6 ± 0.7	9.1 ± 1.1	10.5 ± 2.6	29.6 (<0.05)	2.92 (0.1)
N [mg/100 g]	173.3 ± 11.4	187.2 ± 39.3	265 ± 11.9	263 ± 20.3	75.54 (<0.05)	0.38 (0.55)
P [mg/100 g]	85.4 ± 11.6	74.2 ± 8.1	54.1 ± 6.0	54.2 ± 3.1	64.07 (<0.05)	2.99 (0.1)
	irrigated with TW	irrigated with STWW	irrigated with TW	irrigated with STWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.05 ± 0.02	0.04 ²	< 0.10	< 0.10		
B [mg/kg]	-	-	2.67 ± 0.74	2.48 ± 0.21		
Cd [mg/kg]	0.01 ± 0 ³	0.01 ± 0 ³	0.07 ± 0.01	0.08 ± 0.02	442.65 (<0.05)	2.64 (0.13)
Cr [mg/kg]	0.13 ± 0.09	0.08 ± 0.03	0.17 ± 0.09	0.15 ± 0.03	2.21 (0.16)	0.73 (0.4)
Cu [mg/kg]	1.62 ± 0.23	1.67 ± 0.4	2.08 ± 0.18	1.93 ± 0.24	10.1 (<0.05)	0.2 (0.66)
Fe [mg/kg]	4.73 ± 0.68	4.05 ± 0.91	3.48 ± 0.47	3.79 ± 0.47	8 (<0.05)	0.48 (0.5)
Ni [mg/kg]	0.07 ± 0.03	0.08 ± 0.02	0.27 ± 0.07	0.3 ± 0.08	84.92 (<0.05)	0.78 (0.39)
Pb [mg/kg]	0.18 ± 0.15	0.12 ± 0.13	0.28 ± 0.05	0.23 ± 0.07	5.35 (<0.05)	1.64 (0.21)
Sn [mg/kg]	0.06 ± 0.03	0.06 ± 0.02	0.96 ± 0.15	0.81 ± 0.1	294.86 (<0.05)	3.11 (0.1)
Zn [mg/kg]	2.33 ± 0.26	2.47 ± 0.37	3.90 ± 0.30	3.81 ± 0.42	20.23 (<0.05)	0.02 (0.89)
K [mg/100 g]	224.4 ± 7.1	232.7 ± 16.2	485.3 ± 18.6	476.7 ± 8.8	2084.74 (<0.05)	0 (0.97)
Na [mg/100 g]	5.7 ± 1.4	8.4 ± 2.1	9.1 ± 1.1	9.1 ± 1.9	9.28 (<0.05)	3.61 (0.07)
N [mg/100 g]	173.3 ± 11.4	175.2 ± 16.9	265 ± 11.9	253 ± 15.3	217.97 (<0.05)	0.78 (0.39)
P [mg/100 g]	85.4 ± 11.6	78.2 ± 6.6	54.1 ± 6.0	51.2 ± 1.7	93.65 (<0.05)	2.76 (0.11)

("<LQ": all values below limit of quantification; 1 = only one value above LQ; 2 = only one values above LQ; 3: all value equal to 0.01)

Table 6
Inorganic elements concentration in TW, PTWW and STWW used for potatoes irrigation.

	year 1			year 2			year 3		
	TW	PTWW	STWW	TW	PTWW	STWW	TW	PTWW	STWW
As[μg/l]	<LQ	2.20	3.45	<LQ	3.50	<LQ	<LQ	2.53	7.20
B[μg/l]	139.50	189.00	457.00	149.00	1051.50	143.50	127.67	876.00	511.25
Cd[μg/l]	<LQ	<LQ	<LQ	0.75	1.85	2.00	<LQ	1.18	4.20
Cr[μg/l]	3.05	9.4	3.95	1.00	1.15	1.00	0.266	3.075	27.725
Cu[μg/l]	9.00	2.00	12.00	8.45	23.00	9.50	5.36	20.25	122.28
Fe[μg/l]	<LQ	<LQ	0.04	0.03	0.05	<LQ	<LQ	0.58	0.23
Ni[μg/l]	3.00	4.50	6.05	1.30	7.05	1.35	0.43	12.63	6.95
Pb[μg/l]	<LQ	<LQ	<LQ	<LQ	1.20	<LQ	<LQ	3.50	61.28
Zn[μg/l]	31.50	5.00	139.00	70.50	256.00	89.50	16.13	352.28	334.93
Ca[μg/l]	76.10	70.20	98.70	116.35	105.55	116.40			
K[μg/l]	2.80	7.00	15.70	2.60	10.95	2.85			
Mg[μg/l]	24.15	18.60	23.95	23.95	23.05	23.25			
Na[μg/l]	29.95	72.50	117.25	30.70	91.10	32.25			

of the compounds (Fig. 4). This confirms that the underlying processes driving the variations of inorganic compounds in soils t have to be looked for in the environmental context of the study and in the cropping conditions more than in the irrigation water composition.

3.5. Potatoes crops

The third year, the irrigated potatoes are different by their concentration in Cu, Fe and Zn (Table 8). The F value for these elements is above the critical value.

The potatoes irrigated with and STWW and PTWW contain more Zn which could be related to the concentration in water. Considering Cu, concentration is thirty times higher in PTWW whereas no differences could be measured in the potatoes Cu concentration, irrigated by PTWW.

For other element, no clear relationship could be observed between irrigation water and raw potato chemical quality. Higher the B uptake

by the plant will inhibit Zn uptake (Araújo et al., 2018; Loneragan and Webb, 1993). The trend of the ratio among these elements measured in the tuber confirms the effect of the irrigation water B concentration. Nevertheless, the B contents of the raw potatoes remain in the expected range (6.0–13.0 mg/kg) regardless to the element concentration in irrigation water.

Like in case of the tomato experiments, the potatoes harvested in Year 1 have a different composition than those harvested in Year 3 (Fig. 5). This is confirmed by the results of the 2-ways ANOVA analyses (Table 8)

The variations observed are very important but evolutions of the tuber compositions are similar in the three set of plots, no matter which irrigation water type was applied. The intriguing conclusion is that the inorganic composition of the soil and the raw food seems obviously overridden by external sources. Inorganic compound input may have different sources including fertilisers (Giuffrède López Carnelo et al., 1997), atmospheric dust and aerosols (Cary et al., 2015). Other factors

Table 7

2-ways ANOVA analyse - Element concentration in the soil before the first planting and after harvests of each year (mean for plot cultivated in potatoes according to their type of irrigation).

	Soil before cultivation in year 1		Soil after cultivation in year 3		ANOVA	
	irrigated with TW	irrigated with PTWW	irrigated with TW	irrigated with PTWW	Effect of Year	Effect of Treatment
As [mg/kg]	20.50 ± 0.84	21.00 ± 1.55	36.33 ± 3.27	38.17 ± 4.02	218.29 (<0.05)	1.09 (0.31)
B [mg/kg]	104 ± 14	107 ± 13	88 ± 15	88 ± 19	8.11 (<0.05)	0.08 (0.78)
Cd [mg/kg]	<LQ	<LQ	<LQ	<LQ		
Cr [mg/kg]	127 ± 9	128 ± 8	138 ± 10	139 ± 10	8.07 (<0.05)	0.03 (0.86)
Cu [mg/kg]	50.67 ± 2.73	51.17 ± 3.43	45 ± 4.47	45.33 ± 3.2	16.03 (<0.05)	0.08 (0.77)
Fe2O3 [%]	5.25 ± 0.41	5.33 ± 0.34	4.93 ± 0.5	5.00 ± 0.52	3.14 (0.09)	0.17 (0.69)
Ni [mg/kg]	57.00 ± 5.73	58.33 ± 3.88	66.83 ± 4.49	68.67 ± 5.85	23.85 (<0.05)	0.59 (0.45)
Pb [mg/kg]	26.33 ± 2.88	29.6 ± 4.62	22.6 ± 1.67	26.6 ± 2.07	5.4 (<0.05)	7.62 (<0.05)
Zn [mg/kg]	131.83 ± 4.31	131.5 ± 3.02	117.67 ± 9.27	118.67 ± 9.77	20.93 (<0.05)	0.01 (0.91)
CaO [%]	11.12 ± 1.4	10.70 ± 1.36	9.37 ± 0.21	9.33 ± 0.51	14.16 (<0.05)	0.3 (0.59)
K2O [%]	2.53 ± 0.2	2.6 ± 0.18	2.70 ± 0.17	2.68 ± 0.16	3.02 (0.1)	0.12 (0.73)
MgO [%]	2.53 ± 0.16	2.55 ± 0.14	2.48 ± 0.22	2.48 ± 0.26	0.51 (0.48)	0.01 (0.92)
Na [%]	0.69 ± 0.1	0.65 ± 0.09	-	-		
P2O5 [mg/kg]	3197 ± 390	3283 ± 262	2841 ± 337	3095 ± 242	4.53 (<0.05)	1.76 (0.2)
	irrigated with TW	irrigated with STWW	irrigated with TW	irrigated with STWW)	Effect of Year	Effect of Treatment
As [mg/kg]	20.50 ± 0.84	<LQ	36.33 ± 3.27	38.83 ± 3.31	322.87 (<0.05)	1.07 (0.31)
B [mg/kg]	104 ± 14	110 ± 11	88 ± 15	91 ± 12	11.49 (<0.05)	0.63 (0.44)
Cd [mg/kg]	<LQ	<LQ	<LQ	<LQ		
Cr [mg/kg]	127 ± 9	129 ± 7	138 ± 10	141 ± 9	9.41 (<0.05)	0.42 (0.52)
Cu [mg/kg]	50.67 ± 2.73	51.33 ± 2.42	45 ± 4.47	46.33 ± 2.73	16.73 (<0.05)	0.59 (0.45)
Fe2O3 [%]	5.25 ± 0.41	5.42 ± 0.26	4.93 ± 0.5	5.08 ± 0.41	3.86 (0.06)	0.92 (0.35)
Ni [mg/kg]	57.00 ± 5.73	58.83 ± 1.47	66.83 ± 4.49	69.17 ± 3.97	34.42 (<0.05)	1.47 (0.24)
Pb [mg/kg]	26.33 ± 2.88	25.2 ± 3.96	22.6 ± 1.67	26 ± 2.19	3.06 (0.1)	0.17 (0.68)
Zn [mg/kg]	131.83 ± 4.31	132.17 ± 3.49	117.67 ± 9.27	122 ± 5.48	24.23 (<0.05)	0.89 (0.36)
CaO [%]	11.12 ± 1.4	10.62 ± 0.97	9.37 ± 0.21	9.22 ± 0.3	19.69 (<0.05)	0.84 (0.37)
K2O [%]	2.53 ± 0.2	2.58 ± 0.12	2.70 ± 0.17	2.78 ± 0.12	8.58 (<0.05)	1.13 (0.3)
MgO [%]	2.53 ± 0.16	2.58 ± 0.12	2.48 ± 0.22	2.53 ± 0.2	0.47 (0.5)	0.47 (0.5)
Na [%]	0.69 ± 0.1	0.65 ± 0.09	-	-		
P2O5 [mg/kg]	3197 ± 390	3175 ± 239	2841 ± 337	3089 ± 180	3.3 (0.08)	0.86 (0.36)

("<LQ": all values below limit of quantification; 1 = only one value above LQ; 2 = only one values above LQ)

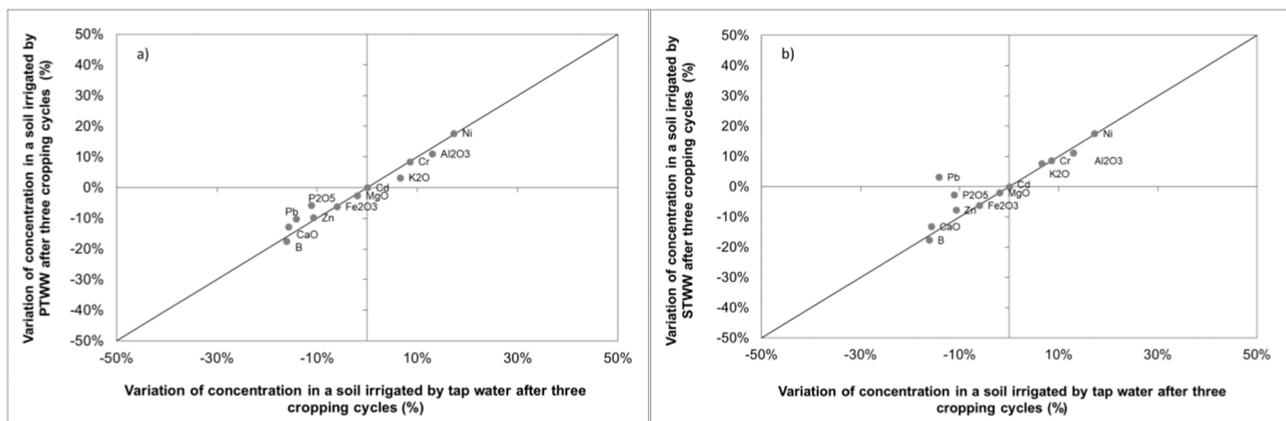


Fig. 4. Comparison of the concentration variation of the inorganic compounds in the soil, under potato cropping, after three cropping cycles. Comparison between tap water irrigation and a) PTWW, and b) STWW.

are related to the irrigation regime (Li et al., 2010), or to the sampling period (Hernández Suárez et al., 2007).

An explanation could be that the edible organs concentrations are not representative of the total plant contamination. The concentration in the edible organs depends on the inorganic compound mobility from the roots to the others organs. Some compounds are very mobile (N, P, K) but others are more static. For instance, Pb is considered to have low mobility within potato plants (Peralta-Videa et al., 2009), which explains why Pb concentrations in soil or in root generally exceed those in tubers (Baghour et al., 2001; Nițu et al., 2019).

It can be furthermore supposed that surface or subsurface drip irrigation will lead to very localised trace element accumulation, driven by geochemical processes in the zone of influence of the dripper (Cary et al., 2015; Guan et al., 2013). Geochemical processes (sorption,

precipitation, complexation, and redox reactions) affect the accumulation of trace metals in environmental systems. Sorption and adsorption to soil particles, especially organic matter and clays, reduce metal mobility by immobilising metals in solid phases. Precipitation, induced by cation reactions with anions like carbonates or sulphides, forms insoluble metal compounds, lowering bioavailability. Complexation with organic or inorganic ligands can enhance the solubility and mobility of metals, increasing their bioavailability. Redox conditions influence the solubility of metals, with reduced forms often being more mobile. In addition, heavy metals can influence each other during the absorption or translocation (Ogugua et al., 2023; Solti et al., 2011). The mean concentrations of heavy metals may vary significantly with the distance from the dripper (Wei et al., 2016).

The quantity of water absorbed by crops impacts, for instance, the

Table 8
2-ways ANOVA analyse - Element concentration in potato tubers for each harvest depending of the water treatment.

	Potatoes cultivated in year 1		Potatoes cultivated in year 3		ANOVA	
	irrigated with TW	irrigated with PTWW	irrigated with TW	irrigated with PTWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.08 ± 0.06	0.05 ± 0.02	< 0.10	< 0.10		
B [mg/kg]			10.31 ± 2.35	6.50 ± 2.07		
Cd [mg/kg]	0.02 ± 0.01	0.02 ± 0.01	0.08 ± 0.02	0.08 ± 0.01	296.05 (<0.05)	0.47 (0.5)
Cr [mg/kg]	0.03 ± 0.02	0.03 ± 0.01	0.13 ± 0.03	0.14 ± 0.04	80.29 (<0.05)	0.04 (0.84)
Cu [mg/kg]	1.74 ± 0.58	2.31 ± 0.32	1.36 ± 0.19	1.21 ± 0.25	17.86 (<0.05)	0.33 (0.57)
Fe [mg/kg]	2.62 ± 0.56	2.87 ± 0.5	4.75 ± 1.3	7.94 ± 1.28	96.67 (<0.05)	13.94 (<0.05)
Ni [mg/kg]	0.37 ± 0.2	0.21 ± 0.2	0.47 ± 0.16	0.54 ± 0.03	0.59 (0.45)	1.51 (0.23)
Pb [mg/kg]	0.05 ²	0.03 ²	0.27 ± 0.131	0.24 ± 0.09		
Sn [mg/kg]	0.18 ± 0.12	0.12 ± 0.11	0.63 ± 0.09	0.67 ± 0.12	3	3
Zn [mg/kg]	2.8 ± 0.23	2.89 ± 0.29	20.57 ± 2.02	32.90 ± 4.58	550.28 (<0.05)	35.3 (<0.05)
K [mg/100 g]	384 ± 30	372 ± 17	459 ± 31	458 ± 14	54.76 (<0.05)	0.51 (0.48)
Na [mg/100 g]			10.79 ± 0.51	11.55 ± 0.95		
N [mg/100 g]	123 ± 25	124 ± 15	247 ± 21	248 ± 18	227.6 (<0.05)	0.07 (0.8)
P [mg/100 g]	148 ± 7	143 ± 9	163 ± 26	150 ± 25	1.1 (0.31)	0.69 (0.42)
	irrigated with TW	irrigated with STWW	irrigated with TW	irrigated with STWW	Effect of Year	Effect of Treatment
As [mg/kg]	0.08 ± 0.06	0.09 ± 0.07	< 0.10	< 0.10		
B [mg/kg]			10.31 ± 2.35	8.62 ± 2.07		
Cd [mg/kg]	0.02 ± 0.01	0.02 ± 0.01	0.08 ± 0.02	0.07 ± 0.03	73.7 (<0.05)	0.14 (0.71)
Cr [mg/kg]	0.03 ± 0.02	0.02 ± 0.01	0.13 ± 0.03	0.14 ± 0.02	125.74 (<0.05)	0.74 (0.4)
Cu [mg/kg]	1.74 ± 0.58	2.05 ± 0.25	1.36 ± 0.19	1.45 ± 0.16	18.44 (<0.05)	5.25 (<0.05)
Fe [mg/kg]	2.62 ± 0.56	2.36 ± 0.25	4.75 ± 1.3	3.64 ± 1.1	14.62 (<0.05)	1.29 (0.27)
Ni [mg/kg]	0.37 ± 0.2	0.51 ± 0.31	0.47 ± 0.16	0.87 ± 0.45	11.43 (<0.05)	1.1 (0.31)
Pb [mg/kg]	0.05 ²	0.31 ¹	0.27 ± 0.131	0.31 ± 0.12		
Sn [mg/kg]	0.18 ± 0.12	0.08 ± 0.05	0.63 ± 0.09	0.48 ± 0.13		
Zn [mg/kg]	2.8 ± 0.23	2.63 ± 0.16	20.57 ± 2.02	32.9 ± 4.58	197.35 (<0.05)	6.47 (<0.05)
K [mg/100 g]	384 ± 30	368 ± 30	459 ± 31	458 ± 14	52.92 (<0.05)	0.38 (0.55)
Na [mg/100 g]			10.79 ± 0.51	11.23 ± 0.84		
N [mg/100 g]	123 ± 25	126 ± 14	247 ± 21	248 ± 29	172.72 (<0.05)	0.02 (0.9)
P [mg/100 g]	148 ± 7	149 ± 4	163 ± 26	153 ± 14	3.57 (0.07)	1.26 (0.27)

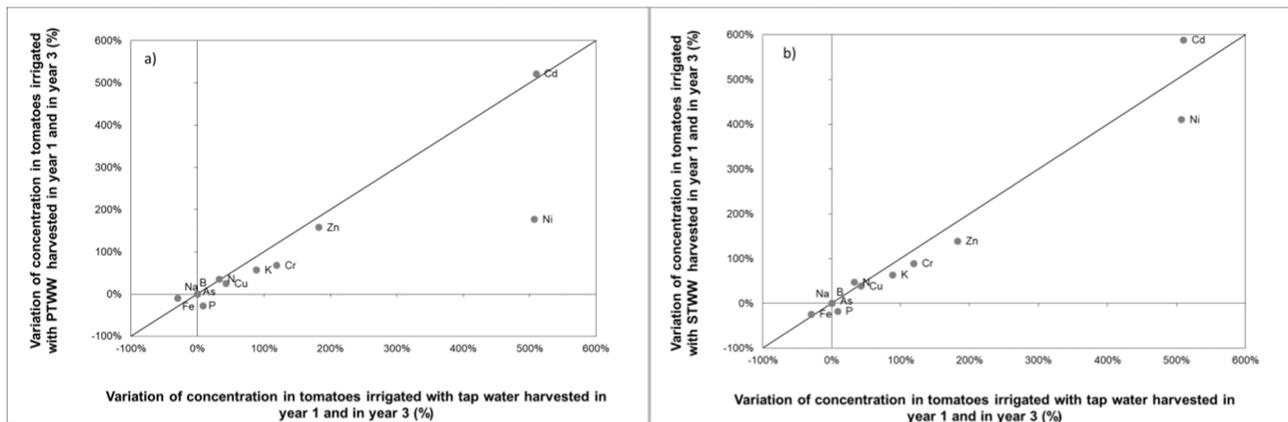


Fig. 5. Comparison of the concentration variation of the inorganic compounds in potatoes after three cropping cycles. Comparison between tap water irrigation and a) PTWW, and b) STWW.

quantity of potassium absorbed (Zhang et al., 2022). The project choice to conduct some plots in full irrigation the first year and in RDI the following year may have had an influence on the absorption even if yield and product quality were comparable (Jensen et al., 2010).

3.6. Potatoes flakes

The potato flakes processed from tubers irrigated during year 3 are different by their content in P, Fe, Pb, Na and Zn (Table 9). The F value for these elements is above the critical value. Yet, evolutions of the flakes concentrations compositions are similar in the three set of plots. The difference between the concentrations in flakes could not be attributed to the irrigation water. For instance, STWW is more concentrated than tap water in Cr during the third irrigation period whereas potatoes flakes from plots irrigated with STWW are less concentrated in this compound.

Due to the industrial processes involved it is difficult to directly link the water constitution and the flake contents. For most of the compounds, the concentrations increase significantly between the raw food and the processed food except for B which is more or less stable. This evolution can be attributed to the processes used for potato flake production (drying/frying). Some previous studies showed that cooking processes have an important impact on inorganic compounds concentrations (Perelló et al., 2008). Depending on the element, the losses during frying can be more or less important (Ersoy et al., 2006). Concerning B, the potato contains different pectins distributed in the cell walls (Bush and McCann, 1999), like RGA II (rhamnogalacturonic acid II), that contain B as a structural part of their molecules (Shi et al., 2017). Even if the skin presents high concentration of B, the potato derivate will still contain a close-to stable B content once the skin is removed. Given the results on raw food and the important alterations caused by food processing, no clear statement can be established.

Table 9

Element concentration in potato flakes for each harvest depending of the water treatment. ("<": below limit of quantification).

	tomatoes puree processed in year 1		tomatoes puree processed in year 3		ANOVA	
	irrigated with TW	irrigated with PTWW	irrigated with TW	irrigated with PTWW	Effect of Year	Effect of Treatment
As [mg/kg]	<LQ	<LQ	<LQ	<LQ		
B [mg/kg]	-	-	5.03 ± 2.01	10.33 ± 4.27		
Cd [mg/kg]	0.09 ± 0.01	0.09 ± 0.01	0.32 ± 0.11	0.31 ± 0.05	83.35 (<0.05)	0.01 (0.92)
Cr [mg/kg]	1.35 ± 0.53	1.22 ± 0.79	1.19 ± 0.49	1.03 ± 0.42	0.56 (0.46)	0.42 (0.53)
Cu [mg/kg]	6.11 ± 0.4	5.56 ± 0.34	7.69 ± 0.78	7.68 ± 0.95	45.8 (<0.05)	1.04 (0.32)
Fe [mg/kg]	25.69 ± 3.97	20.19 ± 3.26	25.41 ± 3.16	30.66 ± 3.77	12.32 (<0.05)	0.01 (0.93)
Ni [mg/kg]	0.97 ± 0.16	1.17 ± 0.5	1.64 ± 0.76	2.55 ± 1.3	9.91 (<0.05)	2.89 (0.1)
Pb [mg/kg]	0.32 ± 0.17	0.19 ± 0.07	1.15 ± 0.56	2.13 ± 0.11	128.74 (<0.05)	12 (<0.05)
Sn [mg/kg]	0.06 ± 0.03	0.1 ± 0.05	0.96 ± 0.15	0.95 ± 0.07	8.21 (<0.05)	0.02 (0.9)
Zn [mg/kg]	17.3 ± 1.2	16.1 ± 1.0	17.9 ± 2.9	13.3 ± 0.8	2.77 (0.11)	17.46 (<0.05)
K [mg/100 g]	750 ± 46	694 ± 65	1785 ± 124	1900 ± 133	763.24 (<0.05)	0.53 (0.48)
Na [mg/100 g]	7.0 ± 2.1	9.5 ± 2.5	22.4 ± 7.9	47.7 ± 20.0	36.25 (<0.05)	9.76 (<0.05)
N [mg/100 g]	1180 ± 95	1156 ± 143	1235 ± 130	1301 ± 83	4.51 (<0.05)	0.19 (0.67)
P [mg/100 g]	706 ± 26	641 ± 49	397 ± 26	406 ± 11	459.76 (<0.05)	4.65 (<0.05)
	irrigated with TW	irrigated with STWW	irrigated with TW	irrigated with STWW	Effect of Year	Effect of Treatment
As [mg/kg]	<LQ	<LQ	<LQ	<LQ		
B [mg/kg]	-	-	5.03 ± 2.01	9.32 ± 3.37		
Cd [mg/kg]	0.09 ± 0.01	0.08 ± 0.01	0.32 ± 0.11	0.37 ± 0.07	93.08 (<0.05)	0.64 (0.43)
Cr [mg/kg]	1.35 ± 0.53	0.85 ± 0.22	1.19 ± 0.49	1.08 ± 0.37	0.03 (0.86)	3.25 (0.09)
Cu [mg/kg]	6.11 ± 0.4	5.38 ± 0.80	7.69 ± 0.78	7.84 ± 0.42	61.88 (<0.05)	1.27 (0.27)
Fe [mg/kg]	25.69 ± 3.97	19.04 ± 1.93	25.41 ± 3.16	26.57 ± 2.22	9.19 (<0.05)	5.26 (<0.05)
Ni [mg/kg]	0.97 ± 0.16	0.80 ± 0.18	1.64 ± 0.76	1.77 ± 0.34	21.52 (<0.05)	0.02 (0.9)
Pb [mg/kg]	0.32 ± 0.17	0.29 ± 0.16	1.15 ± 0.56	1.66 ± 0.67	35.4 (<0.05)	1.69 (0.21)
Sn [mg/kg]	0.06 ± 0.03	0.06 ± 0.02	0.96 ± 0.15	0.81 ± 0.1	2.4 (0.14)	0.55 (0.47)
Zn [mg/kg]	17.3 ± 1.2	15.2 ± 1.6	17.9 ± 2.9	11.2 ± 1.9	4.37 (<0.05)	29.33 (<0.05)
K [mg/100 g]	750 ± 46	676 ± 64	1785 ± 124	1926 ± 180	581 (<0.05)	0.48 (0.49)
Na [mg/100 g]	7.0 ± 2.1	10.7 ± 1.8	22.4 ± 7.9	74.6 ± 19.0	87.23 (<0.05)	43.4 (<0.05)
N [mg/100 g]	1180 ± 95	1096 ± 113	1235 ± 130	1191 ± 108	2.7 (0.12)	1.97 (0.18)
P [mg/100 g]	706 ± 26	625 ± 55	397 ± 26	407 ± 22	348.18 (<0.05)	6.19 (<0.05)

4. Conclusion

This study concludes that small-scale water treatment devices, implemented at farm level and within normal farming practices, can bring down trace metal and nutrient concentrations in irrigation water to levels inducing no harmful variations of inorganic compounds in soils and the edible parts of two different crops (tomatoes and potatoes) after three years of cropping. This study comes to similar conclusions as other experiments performed under comparable conditions in Serbia (Surdyk et al., 2010) and in Crete (Cary et al., 2015).

However, potential contaminations of soil and foodstuff at farm scale may come from multiple and not necessarily controlled sources other than irrigation water. An interesting lesson from this study is therefore to show how difficult it will be to define thresholds of inorganic compounds in irrigation water since normal farming practices (full, deficit and drip irrigation regimes, fertilisation, pesticide treatment) already lead to highly variable soil concentrations.

This study provides a real-world impact assessment of low-quality irrigation water with no tangible effects within a concentration range that we esteem rather representative of TWW under the prevalent legal framework in the EU. Compared to previous studies with proven impacts on soil and crop quality (Aslam et al., 2023; Singh et al., 2010), our TWW concentrations are relatively low (except B and Zn concentration which are among the highest concentration published) but our study confirms results in similar water quality ranges (Arora et al., 2008, Libutti et al., 2018). Whereas it may seem reassuring that in this range effects are small compared to those of other factors outline above, our study does not allow definitive conclusions on the long-term effects of irrigation with treated wastewater since those seem to depend on the initial concentration of metal in irrigation water. However, defining clear thresholds is economically important since in the actual context irrigation with high quality water seem more and more jeopardised. Furthermore, new pollutant classes emerge, following rapid technical and societal evolutions. While using the experimental framework developed by our study, further research needs to include pollutants as

pharmaceutical residues, microplastics and industrial compounds (PFAS) contained in municipal wastewater.

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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Data availability

Data will be made available on request.

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