Agricultural Water Management 155 (2015) 87–99

Contents lists available at ScienceDirect

Agricultural Water Management

journal homepage: www.elsevier.com/locate/agwat

Short-term assessment of the dynamics of elements in wastewater irrigated Mediterranean soil and tomato fruits through sequential dissolution and lead isotopic signatures

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ARTICLE INFO

Article history:
Received 11 November 2014
Received in revised form 13 March 2015
Accepted 16 March 2015

Keywords:
Trace metals
Salinity
Boron
Crete
Drip irrigation
Soil mapping

ABSTRACT

To fight against sanitary risks due to the use of raw or insufficiently treated wastewaters, an irrigation system combining a farm-scale decentralized wastewater treatment and an improved drip irrigation management was developed. The whole soil-water-plant system was monitored on an experimental tomato field in Crete to assess the potential element accumulation due to drip irrigation with secondary treated wastewaters during 3 years. Although a decrease of the majority of element average concentrations (Mn, As, Cr, Cu, Pb, Zn and B) was observed in irrigated soils possibly due to crop export, increasing amounts of elements, especially for As, Cd, Zn, Cu, Ni and Na were measured in the bio-accessible and mobilizable fractions of the soil. Moreover, we show that surface drip irrigation can lead to very localized accumulation of trace metals driven by geochemical processes as pointed out by soil mapping. Transient but significant Na and B contents can also develop under the emitter in the soil fractions during secondary treated wastewater irrigation. Pb displayed no spatial relation with the drip emitter and its main sources were petrol-Pb aerosols and Saharan dusts. The trace metal contents and salinity indicators in crops stayed below the regulatory thresholds. Sequential dissolutions combined with the Pb isotopic tool traced element mobility and dynamics in an irrigated soil at short term, and were useful to distinguish between medium-term experiment at bulk soil scale and short-term impacts of secondary treated wastewaters irrigation at the drip emitter scale.

1. Introduction

The sustainable use of water is a priority for agriculture where, as in Mediterranean regions, water resources are insufficient to meet all needs (Angelakis et al., 1999). In islands like Crete, summer tourism increases fresh water needs further during the period where irrigation water demand is already at its highest. A way to keep water available for agriculture without reducing water supply for other uses is to reuse wastewater (Toze, 2006). When included in an integrated approach of water resources management, wastewater is designed to compensate for overall water scarcity but also helps to solve one of the main problems of the Mediterranean basin and Middle East region, i.e., the annual variations of water availability and the related need for water storage. Nevertheless, environmental and health risks are linked to wastewater reuse for agricultural purposes, requiring imperatively adequate treatment and water storage (Asano and Levine, 1996). Depending on the quality and quantity of the water being spread, wastewater irrigation of crops during decades can lead (1) to the accumulation of metallic trace elements in the underlying soil and plants, e.g., around Paris or Mexico (Ahumada et al., 1999; Cajuste et al., 1991; Dere et al., 2006, 2007; Huerta et al., 2002; Lucho-Constantino et al., 2005) or elsewhere (Gupta et al., 2012; Leal et al., 2009; Sharma et al., 2007) and (2) to soil salinization especially under modern and intensive agricultural system in arid and semi-arid areas because almost all types of effluents are moderate to highly saline, e.g., in Crete (Chartzoulakis and Psarras, 2005; Paranychianakis and Chartzoulakis, 2005). On one hand, metals are a concern to human health because they tend to bioaccumulate in the food chain (Järup, 2003), and on the other hand, soil salinization affects the physical and chemical properties of soils. Furthermore, the combination of water saving strategies based on the use of

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http://dx.doi.org/10.1016/j.agwat.2015.03.016
0378-3774/

reclaimed water can result in negative impacts on the plant production, on the availability of water in the soil and on the growth pattern of the plant rooting system. However, the concentrations of metals in treated wastewater being generally low, their potential impact on plant production and on the soil are generally taken into account after a long-term period on wide scale trials, neglecting possible local and short-term effects. An appropriate knowledge of the effects of irrigation on the spatial and temporal variability of the chemical composition of the irrigated soils in the early periods of irrigation is thus essential to better predict long-term trends, especially for metals, sodium and boron.

Metals in the soil solution are subject to mass transfer out of the system by leaching to groundwater, plant uptake or volatilization. At the same time metals participate in chemical reactions with the soil solid phase. The concentration of metals in the soil solution, at any given time, is governed by a number of interrelated processes, including inorganic and organic complexation, oxidation reduction reactions, precipitation/dissolution reactions and adsorption/desorption reactions (Kabata-Pendias, 2004). The ability to predict the concentration of a given metal in the soil solution depends on the accuracy of changes in soil environmental conditions over time, such as the degradation of the organic matter, changes in pH, redox potential or soil solution composition, due to various remediation schemes such as irrigation with treated wastewaters. The extent of vertical contamination is related to the soil solution and surface chemistry of the soil matrix. Speciation may also evolve over time. To characterize the metal fractions in soils and sediments and predict their mobility and bioavailability (Tessier et al., 1979) especially in soils irrigated with wastewaters, sequential extraction techniques are widely used, although their efficiency and specificity are often discussed and improved (Dong et al., 2000; Khanmirzaei et al., 2013; Neaman et al., 2004; Trolard et al., 1995). To distinguish between the bioavailable fraction and the mobilizable metallic fraction to better focus on plant uptake, sequential extractions of soil fractions combined with lead isotopic composition determination are used. Lead isotopic composition is a powerful tool to distinguish between sources of Pb in the soil and to better understand the Pb behaviour and infiltration rates (Bacon et al., 2006; Emmanuel and Erel, 2002; Erel et al., 1997; Teutsch et al., 1999, 2001).

In this context, advanced irrigation systems using primary or secondary treated wastewater from water treatment plants and combining specifically designed local treatment on farm level with surface and subsurface drip irrigation and irrigation strategies have been tested in several hydro-climatic contexts (Crete, Italy, Serbia, China) on experimental irrigated tomato and potato fields (Battilani et al., 2010; Li et al., 2010; Surdyk et al., 2010). To better evaluate the short-term impacts of secondary treated wastewater (STWW) irrigation on the soil and fruit quality, an original methodology to study the soil-water-plant system was developed. We used a geochemical approach to assess the magnitude and trends of the soil and tomato quality under surface and subsurface drip irrigation during medium-term use of treated municipal wastewater in Crete, by focusing on trace metals and salinity. We combined sequential extractions of the soil fractions, determination of Pb isotopic composition, and a detailed study of element distribution around drip emitters with soil mapping.

2. Materials and methods

2.1. Site characteristics

The experimental site was located in the Prefecture of Chania in Crete (35°28′39″N and 24°2′34″E). The annual mean rainfall is 740 mm and from May to August, the last 10 years mean temperature in the area is 23 °C, with a mean rainfall of 4 mm per month for these summer months. In normal years, the mean annual potential evapotranspiration for the area is 1450 mm (Region of Crete, 2001). The field site soil is an Alfisol, well drained over its whole depth. The substratum is constituted by Miocene sediments and outcrops close to the study site.

Within the experimental area (approximately 120 m × 20 m), the grain size distribution shows a predominance of the sand fraction (42–66%) together with a clay fraction (14–42%) and a loamy fraction (14–26%). Principal aluminosilicates are feldspars (microcline, plagioclase) and micas, and argillaceous minerals (illite, kaolinite). Minor components as spinel and diopside were also detected. We observed a spatial NE–SW gradient of soil mineralogy at field scale mainly linked to the relative abundance of clays and iron oxides, with a higher CEC (10 meq) and lower porosity. The Fe_2O_3 and MnO contents varied respectively from 2.7% to 6.4% and 0.07% to 0.13%. Organic matter (OM) was concentrated in the uppermost 25 cm (0.6–1.47%). In the lower investigated horizon (25–50 cm), the OM contents ranged from 0.27% to 0.74%. The carbonate content was inferior to 2.2%. Porosity was around 36.4 ± 1.6%. pH was close to neutral, and varied slightly around 6.5 (±0.09). Electrical conductivity was low (0.3 dS/m). The soil is more reddish and clayed at the South-Eastern lower part of the field array of plots.

2.2. Water treatment system

Tomatoes were irrigated with secondary treated wastewater (STWW) provided by the municipal treatment plant of Chania city. STWW were stored on-site in four 5 m^3 tanks and underwent complementary purification, before irrigation, through a specifically developed farm-scale on-site treatment system. The on-site treatment by the modular SAFIR field treatment system (FTS), which was a simple and robust local tertiary treatment, included a specially designed sand filter (produced by Netafim, Israel), a heavy metal removal unit, based on heavy metal adsorption to granular ferric hydroxide and a UV treatment (Battilani et al., 2010). It is an innovative process never applied before to produce functional water to irrigate food crops. As trace metal and metalloid concentrations in the input STWW were found highly variable and low during year 1, they were stabilized by a spike with As, Cd, Cr, Cu and Pb for irrigation during year 2 and 3 to better constrain experimental conditions. A concentrated solution of metal salts was added proportionally to the input wastewater flow so that the following constant target concentrations before the sand filter were obtained: 20 μg/L for As, 5 μg/L for Cd, 100 μg/L for Cr, 200 μg/L for Cu and 100 μg/L for Pb.

2.3. Experimental design and crop management

Treatments were set up by combining two different water sources (fresh water from the irrigation network of Chania Prefecture as the reference treatment and secondary treated wastewater from the local wastewater treatment plant) and two irrigation techniques (surface and subsurface irrigation, Table 1). Each treatment was replicated three times, with each replication (plot) including 100 tomato plants in five rows, occupying a surface area of 50 m^2. Plots were randomly distributed in a randomized block design. Crop management was identical in all plots. The field was ploughed in December, planting took place in April after surface tillage of the soil, and harvest started from mid- to late June and was completed in mid-August. Plants were irrigated by drip lines (Netafim, Israel) at 0 and 15 cm soil depth (for surface and subsurface irrigated plots respectively), starting right after planting and continued until mid-August. Emitters were installed every 50 cm and the plant was located at 25 cm from the drip. One emitter per plant supplied 1.6 L