Science of the Total Environment 584–585 (2017) 595–602

Contents lists available at ScienceDirect

Science of the Total Environment

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

Effects of residual disinfectant on soil and lettuce crop irrigated with chlorinated water

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HIGHLIGHTS

- Halogenated compounds measured in soil and crops irrigated with chlorinated water
- Role of soil organic fraction evidenced pot trials on sandy and silty-clay soils
- Roots damage found to be positively correlated to residual chlorine or chloramine.
- Crops on sandy soil found more sensitive to free chlorine than silty-clay soil.
- Silty-clay soil shows a long-term memory effects than sandy soil.

GRAPHICAL ABSTRACT

Lettuce plants grown on silty-clay soil and watered with increasing free chlorine concentrations in the irrigation water.

ARTICLE INFO

Article history:
Received 11 October 2016
Received in revised form 13 January 2017
Accepted 13 January 2017
Available online 24 January 2017

Editor: D. Barcelo

Keywords:
Irrigation
Tertiary treated wastewaters
Vegetable crops
Residual chlorine
EOX

ABSTRACT

The accidental or continuous release of residual chlorine in water reclaimed for irrigational purposes could compromise the crop yield and increase the load of toxic organo-halogenated compounds, posing additional risks for environment and human health.
This study was aimed at assessing the consequences of using chlorinated water for irrigating lettuce crops grown in pots with two different types of soil.
The results show that the accumulation of extractable organo-halogenated compounds (EOX) in soil, roots and leaves is directly related to the chlorine concentration in the irrigation water. The accumulation of EOX in sandy soils is not significant, while it reached up to 300% of the control in the silty-clay soil, demonstrating that the phenomenon is linked to the organic matter content in the soil. The accumulation of EOX in the soil appears to play a significant role in subsequent bioaccumulation in cultures irrigated with tap water (long term memory effect). Chloramines also demonstrated to have similar impacts as the free chlorine from hypochlorite. The consistent bioaccumulation of 400–700 $\mu g_{Cl}\ kg^{-1}$ of EOX in the leaves of crops irrigated with just 0.2 $mg_{Cl}\ L^{-1}$ of residual chlorine, as compared to levels below the detection limit of 75 $\mu g_{Cl}\ kg^{-1}$ in the control crops, evidences the potential impact on food chain and human health.

1. Introduction

Municipal wastewater can be used for irrigation, representing an attractive alternative to conventional water resources in arid and

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http://dx.doi.org/10.1016/j.scitotenv.2017.01.083

semi-arid countries. However, the irrigational reuse of municipal effluents requires appropriate treatments aimed at avoiding or minimizing both hygienic and agronomic problems.

Tertiary treatments, including flocculation, filtration and disinfection, are typically used to contain microbial risks to human health (Stanfield et al., 2003). The current regulations consider disinfection as an important element of wastewater treatment when necessary to protect public health (WHO, 1989; U.S. E.P.A., 1992). Sodium hypochlorite (NaOCl) is the most widely used disinfectant in water and wastewater treatment plants, thanks to its bactericidal and bacteriostatic properties, as well as to its relatively low cost (Crebelli et al., 2005; Norton-Brandão et al., 2013). Numerous experiences indicate about 10 mg_{Cl} L^{-1} and 30 min as the optimal dosage and contact time to reduce the content of pathogens below the strict reuse limits of the Italian law (Stanfield, 1996; Lazarova et al., 1998). Post-dechlorination treatments are usually introduced to bring the residual chlorine concentration below the prescribed limit (e.g. 0.2 mg_{Cl} L^{-1} according to the Decree of the Italian Ministry for Environment, 2003), minimizing the impact on the receiving body.

While the problems related to microbial contamination have been widely studied and considered close to a solution, those linked to the presence of organic and inorganic chemicals in the treated wastewater are far from resolution (Guide ATV, 1993; Fatta-Kassinos et al., 2011; Kalavrouziotis et al., 2011). Many risks related to the reuse of wastewater for irrigation purposes can be essentially linked to two factors: the first is the contamination of agricultural soils with consequent salinization of the soil, phytotoxic effects and/or contamination of the food chain, and the second is the contamination of subsurface water (Leoni and Fabiani, 1985; Toze, 2006; Kalavrouziotis et al., 2015).

Many national and international organizations are in the process of defining the concept of hazardous substances and regulating their use and release in the environment. In this sense, the Directive 2000/60/EC of the European Parliament established a framework for community action in the field of water policy, and defined “hazardous substances” as substances or groups of substances that are toxic, persistent and liable of bioaccumulation (Contreras Lopez, 2003).

Bioaccumulation is a selective process by which a chemical is concentrated in an organism in quantities greater than the surrounding medium (Emmanuel et al., 2005). Bioconcentration results when the intake far exceeds the clearance rate by an organism. Biomagnification is a descriptor of a process by which the chemical increases its concentration between steps in the food ladder in the environment. The difference of these terms has been stressed in a recent workshop on bioaccumulation by aquatic organism (Bartell et al., 1998). Whereas bioconcentration considers uptake from water; bioaccumulation includes uptake from water, sediment, food and suspended particles (Contreras Lopez, 2003). The uptake, storage, and release of chemicals by the vegetation are also critical components of the global cycling of persistent organic pollutants (Dalla et al., 2004). A knowledge of the uptake and degradation of organic chemicals by plants is also an essential element of any phytoremediation application, and a prior understanding of these processes is implicit in decision-support tools such as the U.S. Environmental Protection Agency (U.S. EPA) online Phyto Decision Tree (ITRC, 2009).

Nontoxic organic compounds in wastewater can be transformed into potentially toxic chlorinated organic compounds when chlorine is used for disinfection purposes. From the chemical point of view, the higher the residual chlorine the greater is the number of by-products that are formed (U.S. E.P.A., 2012). Disinfection by-products from the chlorination of drinking water and wastewater are subjects of continuous investigations worldwide, and many experts in this field have expressed their concerns about the consequences of their release in the environment. Yet, most of the research conducted so far focused primarily on the carcinogenic and mutagenic properties of wastewater chlorination by-products as a public health issue (Akande et al., 2010). Although actual data on the persistence of these by-products on food crops are lacking, their presence should not limit the use of reclaimed water for crop irrigation and further research is required to assess possible issues related to bioaccumulation and transfer to the food chain.

Very little is known about the consequences of the use of chlorinated effluents, although it is generally accepted that the residual chlorine is toxic for sensitive crops already at concentrations of 0.05 mg_{Cl} L^{-1} (U.S. E.P.A., 2004). Hazardous compounds similar to the disinfection by-products can be formed into the soil, in absence of de-chlorination treatments or in case of accidental releases prior to the reuse of the treated wastewater. In these cases, the excess chlorine left in the reused water will react with the organic matter in the soil, leading to the formation of organo-halogenated by-products (OX) that may have genotoxic effects and adverse impacts on both environment and humans (Verlicchi and Masotti, 2002; Crebelli et al., 2005). The principal pathways for the exposure of humans to effluent-born toxic organic compounds through the uptake and transfer to edible portions of the plants, and subsequent consumption by farmed animals and humans were assessed by Collins et al. (2007). It has also been evidenced that the possible accumulation of chloride resulting from the reuse of chlorinated wastewater may turn into increased mobility and bioavailability of heavy metals in the soil, with transfer to plant leafs and food chain (Kalavrouziotis et al., 2011).

The goal of this work was to verify formation and accumulation of organo-halogenated compounds in soil, as well as their availability, uptake and accumulation in the irrigated crops. These organo-halogenated species may affect physiological plant parameters such as photosynthesis and plant growth, bioaccumulation and distribution of OX in plant tissues (Akande et al., 2010). In order to evidence the effects directly related to the interaction between residual chlorine, organic matter in the soil and crops, rather than that of disinfection by-products, chlorinated tap water has been adopted for the irrigation of lettuce crops grown in pots. We also propose to monitor the accumulation of organo-chlorinated compounds by measuring the extractable fraction (EOX, extractable organic halogens) of all organo-halides present in the soil, because this fraction is expected to have higher mobility and to pose the greatest hazard. This paper attempts to highlight a possible risk that has received less attention by past investigators, such as the potential effects of escalating levels of organo-halogenated by-products on quality, performance and overall agricultural productivity of selected test crops.

2. Materials and methods

Before this study, experimental field tests were carried out on lettuce and fennel crops, irrigated with treated municipal wastewater disinfected by sodium hypochlorite (Lonigro, 2006). The residual chlorine values were comprised between 0.5 and 10 mg_{Cl} L^{-1}. Crops showed chlorosis, leaf necrosis, low production and accumulation of halogenated by-products (Lonigro and Rubino, 2008; Lonigro and Rubino, 2010).

In order to further investigate the effects of residual chlorine and toxic by-products from its reaction with the organic matter in the soil, three different trials were conducted on lettuce crop grown in PVC pots and irrigated with water containing increasing concentration of residual chlorine. The PVC pots had max diameter of 0.25 m, height of 0.24 m and volume of 5200 cm^3. The pots were placed under a glass canopy, to avoid the possible dilution effect of rainwater.

Two different soils were compared in a first trial, a sandy and a silty-clay soil (the same of the tests field). The chemical-physical characteristics of these soils are summarized in Table 1. Different test conditions were compared: a control (C) irrigated with tap water (residual chlorine below the limit of detection of 0.04 mg_{Cl} L^{-1} and electrical conductivity of 0.36 dS m^{-1}) and 3 pots irrigated with tap water added with 0.2, 10 and 40 mg_{Cl} L^{-1} of residual chlorine, respectively (max electrical conductivity of 0.50 dS m^{-1}). The experimental design was a randomised block with three replications (Fig. 1).