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Protecting effect of recycled urban wastes (sewage sludge and wastewater) on ryegrass against the toxicity of pesticides at high concentrations

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ABSTRACT

Degraded landscapes, like those from abandoned mine areas, could be restored by revegetating them with appropriate plant species, after correction for acidity and improvement by adding exogenous organic material. Application of urban wastes to large areas of derelict land helps in the sustainable development of this landscape. However, the development of plant species in these soils could require in the future the management of possible pests or diseases by pesticide applications which could also affect plant yield. Therefore, ryegrass (*Lolium perenne* L.) was planted in a limed soil from the mining area of Riotinto (SW Spain), using an indoor pot experiment and the effects of amendment with sewage sludge, as well as irrigation with urban wastewater on plant uptake of the insecticide thiacloprid and the fungicide fenarimol were examined. Ryegrass biomass was reduced up to 3-fold by pesticide application. Fenarimol residues were the highest in soil, while those of thiacloprid were lower in soil and higher in ryegrass. Addition of sewage sludge and irrigation with wastewater led to a reduction of pesticide translocation to the aerial plant parts, representing a lower hazard to ryegrass quality grown in this mine soil.

1. Introduction

Mine soils suffer from high metal concentration, sometimes above the legal guidelines. For instance the Riotinto mine area has high heavy metal load making this environment inappropriate for plant establishment and growth, though different plant species have shown their ability to develop under these extremely harsh conditions (Trigueros et al., 2012; Abreu et al., 2012). Due to the degradation of this landscape, one of the strategies to cope with this situation has focused on the revegetation of the mine areas, because this will contribute with beneficial direct or indirect effects, such as erosion control, site restoration or carbon sequestration.

For successful revegetation, various measures are usually introduced oriented to the correction of soil acidity through liming or addition of exogenous organic amendments to improve soil OC and fertility. Different organic amendments of agricultural or industrial origin have already demonstrated the improvement of soil quality (Mingorance et al., 2014) allowing the development of healthy plant species (Wang et al., 2012). They also help in the development of soil aggregation, representing an early step of soil reclamation in mine waste deposits and bridging the physical and biological properties of soil systems. Among them, land application of treated sewage sludges helps to maintain a sustainable environmental management by recycling these low-cost wastes, reducing their disposal and returning organic material, trace elements, moisture and nutrients to the soil (García-Gil et al., 2004; Mingorance et al., 2014).

However, the development of plant species in these soils could require in the future the management of possible pests or

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diseases by using pesticides which could also affect plant yield. The presence of organic contaminants in the soil together with heavy metals from past metallurgical activities, may impair the development of the vegetal species planted for revegetation, as well as incorporate residues of the organic pollutants into the plant. The degree of plant uptake will depend on the plant species but also on the properties of pesticides (Yu et al., 2009; Jiang et al., 2011), because the movement of non-ionic organic compounds into the roots can be considered a partition between liquid and solid phases (Sicbaldi et al., 1997) related with pesticide polarity.

In reclamation of mine soils, use of organic amendments, such as sewage sludges, has been focused on the management of inorganic pollutants, i.e. heavy metals (Alvarenga et al., 2009a). In other assays with plants, addition of organic amendments from different sources (biochars, manure, sludges) has been reported to enhance soil retention of organic pollutants (Yu et al., 2009; Hilber et al., 2009; Jiang et al., 2011), reducing plant uptake. Besides, amendments with high microbial activity, like manure or sewage sludge, apart from the benefit of recycling, may also enhance degradation processes, which further reduce pollutant plant uptake (Jiang et al., 2011; Wang et al., 2012).

In addition, in arid or semiarid regions shortage of fresh water restricts agricultural production or else plant development and yield production may be compromised. Furthermore if water is used for irrigation of a degraded soil which in principle will not be devoted to agricultural production, the selection of marginal water, such as treated urban wastewater, will provide a cost-effective and available alternative for irrigation of this impaired soil (Qadir et al., 2007). However, treated wastewater has been shown to modify in some cases the behaviour of pesticides in the soil ecosystem (Rodríguez-Liébana et al., 2011, 2014). Finally, though wastewater contains higher levels of salts and dissolved organic matter than fresh water, which may alter plant performance and yield quality (Sopper and Kardos, 1972; Bernstein et al., 2009), no reference on the effect of wastewater irrigation on pesticide plant uptake could be found in the literature.

The novelty of this work lies on the use of sewage sludge in mine soils oriented to reduction of environmental hazard by organic pollutants and irrigation with wastewater, the latter rarely considered to date in revegetation strategies of mine soils and never included, to our knowledge, in pesticide plant uptake. Perennial ryegrass, *Lolium perenne* (L.), which adequately develops in acid mine soils (Alvarenga et al., 2009a; b; Mingorance et al., 2014), and which has been also proposed as an appropriate species to remediate soils co-contaminated by heavy metals and organic pollutants (Chigbo and Batty, 2013), was selected as plant model species. The aim of the present work was focused on the effects of sewage sludge and wastewater treatments on plant biomass production and on the protection of plant against the toxic effect of two pesticides (an insecticide and a fungicide) with different chemical–physical properties.

2. Materials and methods

2.1. Pesticides

High purity standards (≥98%, Dr. Ehrenstorfer, Germany) of thiacloprid (THC), a nicotinoid insecticide and of the pyrimidine fungicide fenarimol (FEN) were used without further purification. Their octanol/water partition coefficients (log K_{ow}) are 1.26 and 3.69, and their solubility in water 185 and 13.7 mg L^{-1}, respectively (Tomlin, 2003). Stock pesticide solutions were prepared at 1000 mg L^{-1} in acetone.

2.2. Soil, sewage sludge, liming agent and water properties

Soil was collected from the upper layer of an acid mine waste, in the Riotinto mining area (37° 42′ 4.5″ N 6° 33′ 35.1″ W), located in the Iberian Pyrite Belt, which includes one of the largest deposits of pyrite (FeS_2). According to X-Ray Fluorescence analysis SiO_2 together with Fe and Al oxides represent more than 80% of the soil mineralogical composition (Rodríguez-Liébana et al., 2013). It is a very acid sandy loam soil (pH 2.4), with high electrical conductivity (EC, 1.3 dS m^{-1}) (both at 1/2.5 ratio, w/v) and low organic carbon (OC, 1.4%) content. The content of some potential hazardous metals (mg kg^{-1}) (As, 3951; Cd, 13; Cu, 694; Pb, 3976) is above the local guidelines (Mingorance et al., 2014).

The Nerva mine soil (<8 mm) was limed with 1.8% Carbocal, a residue from the sugar industry (Azucarera Ebro) which contains 83.4% $CaCO_3$. The stabilized sewage sludge (SSL), from the wastewater treatment plant of Seville (SW Spain), had an OC content (%) of 22.03 ± 2.94, pH 6.96 ± 0.01 and EC (1/10 ratio, w/v) 1.6 ± 1.2 dS m^{-1}.

Irrigation was performed with distilled water (DW) [pH 5.3 ± 0.3, EC 5.0 ± 1.3 μS cm^{-1}, dissolved OC (DOC) 0.19 ± 0.03 mg C L^{-1}] or wastewater (WW) [pH 8.7 ± 0.3, EC 1063 ± 88 μS cm^{-1}, DOC 21.3 ± 2.8 mg C L^{-1}]. Besides, as reported by the treatment plant, WW also contained 27.3 mg L^{-1} total N and 3.8 mg L^{-1} total P, had a chemical oxygen demand of 51 mg L^{-1} and a biological oxygen demand <13 mg L^{-1}.

2.3. Pot experiments

Limed soil was thoroughly mixed with the organic amendment (SSL at 2%) in a mechanical shaker for 4 h. The SSL dose was selected on the basis of a previous screening with three different plant species (Mingorance et al., 2014), indicating that *L. perenne* was a species easy to cultivate with a short cultivation period and that SSL at 2% promoted plant biomass without affecting seed germination and mortality.

The experiment was carried out in the greenhouse of the University of Seville, with an average temperature of 21.8 °C and relative humidity of 67%. An indoor system was used using plastic pots containing 250 g soil. The treatments ($n = 4$) consisted in non-amended soil irrigated with DW or WW (SSL0-DW and SSL0-WW) and sludge-amended soil at 2% with DW or WW irrigation (SSL2-DW and SSL2-WW). Additionally two treatments without pesticides (SSL0-DW and SSL0-WW) were also included to evaluate the effect of pesticides on biological plant parameters. In order to reduce soil compactness, 40 g of glass beads (4 mm in diameter) were mixed in each pot. Eleven ryegrass seeds were grown directly on each pot and irrigated every 2 days with DW for a week. Then the pots corresponding to treatments with pesticides were added with 5 mL of a solution containing a mixture of THC and FEN at 200 μg mL^{-1} in DW followed by 5 mL DW or WW, depending on the treatment. Pesticide dose corresponds to 2 L a.i. per ha, which is approximately 3 times the recommended dose for THC. This would allow estimating a worse case scenario and quantifying pesticide residues at the end of the experiment.

Afterwards irrigation, necessary to maintain soil water holding capacity and without effluence from the bottom of the pots, was carried out by adding 10 mL water (DW or WW), every two days for 27 days.

Several parameters were measured in plants at the end of the experiment in all the treatments: germination and mortality rates, biomass production (dry weight), shoot length and relative water content. In those treatments containing pesticides, a set of soil and plant subsamples was immediately frozen and kept at their water content at −18 °C, until the measurement of soil dehydrogenase activity and the determination of pesticide residues in soil and plant.