



Arsenic distribution in soils and rye plants of a cropland located in an abandoned mining area



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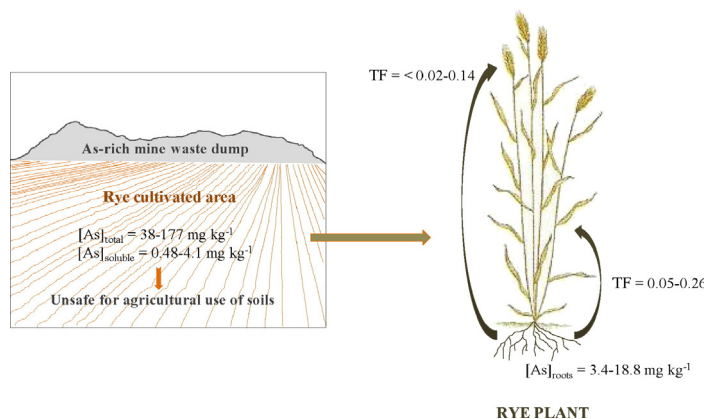
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HIGHLIGHTS

- Environmental assessment of a rye cultivated area impacted by past mining activities.
- Soil As contents exceeded the recommended safe limits for agricultural use of soils.
- Soil soluble As concentrations attained high levels (0.48–4.1 mg kg⁻¹).
- As was mainly distributed in rye roots with limited translocation to shoots and grains.
- Rye shoot and grain As contents meet the prescribed standards of feed and food safety.

GRAPHICAL ABSTRACT



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ABSTRACT

A mining impacted cropland was studied in order to assess its As pollution level and the derived environmental and health risks. Profile soil samples (0–50 cm) and rye plant samples were collected at different distances (0–150 m) from the near mine dump and analyzed for their As content and distribution. These cropland soils were sandy, acidic and poor in organic matter and Fe/Al oxides. The soil total As concentrations (38–177 mg kg⁻¹) and, especially, the soil soluble As concentrations (0.48–4.1 mg kg⁻¹) importantly exceeded their safe limits for agricultural use of soils. Moreover, the soil As contents more prone to be mobilized could rise up to 25–69% of total As levels as determined using (NH₄)₂SO₄, NH₄H₂PO₄ and (NH₄)₂C₂O₄·H₂O as sequential extractants. Arsenic in rye plants was primarily distributed in roots (3.4–18.8 mg kg⁻¹), with restricted translocation to shoots (TF = 0.05–0.26) and grains (TF = <math>< 0.02-0.14</math>). The mechanism for this excluder behavior should be likely related to arsenate reduction to arsenite in roots, followed by its complexation with thiols, as suggested by the high arsenite level in rye roots (up to 95% of the total As content) and the negative correlation between thiol concentrations in rye roots and As concentrations in rye shoots ($|R| = 0.770$; $p < 0.01$). Accordingly, in spite of the high mobile and mobilizable As contents in soils, As concentrations in rye above-ground tissues comply with the European regulation on undesirable substances in animal feed. Likewise, rye grain As concentrations were below its maximum tolerable concentration in cereals established by international legislation.

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1. Introduction

Environmental contamination with arsenic is a matter of great concern due to the serious adverse effects of this trace element on living organisms, especially on mammals (Kabata-Pendias and Mukherjee, 2007). Arsenic toxicity importantly depends on its chemical forms; in general, inorganic species are more toxic than organic ones (Ng, 2005; Sharma and Sohn, 2009). Chronic exposure to inorganic As can cause in humans skin lesions, hematological disorders, immunological, neurological and reproductive alterations, cardiovascular disease, and cancer development (Rahman et al., 2001; Mandal and Suzuki, 2002; Khan et al., 2003).

Increased As levels in the environment can take place by both geogenic and anthropogenic processes. Human activities emitting As mainly include coal/oil combustion, mining, smelting, pesticide production and application, manufacture of alloys, electronics and glass/ceramics, and municipal solid waste incineration (Alloway, 1995; Henke, 2009). Of them, mining activities, although not being the largest source of As into the environment, are considered as one of the most highly polluting.

Humans are exposed to As primarily from air, food, and water (Mandal and Suzuki, 2002). Except for occupational exposure, oral ingestion represents the main route for As intake (Mandal and Suzuki, 2002; Kabata-Pendias and Mukherjee, 2007; Sharma et al., 2014). In this regard, severe poisoning episodes due to As exposure of geogenic origin in drinking water have been reported in different regions of the world (Tseng et al., 1968; Das et al., 1996; Mukherjee and Bhattacharya, 2001; Dogan et al., 2005; Pokhrel et al., 2009; Nicolli et al., 2012). Apart from contaminated drinking water, the consumption of crops grown in areas polluted with As can also result in an important threat to human health. Among crops, cereals are considered the main contributors to dietary inorganic As intake (Schoof et al., 1999; D'Amato et al., 2011; Sharma et al., 2014). Many studies on the As uptake, translocation and accumulation characteristics of cereal crops have been performed under both controlled and field conditions. Most of these studies have been focused on those cereals with the highest production worldwide, namely corn (e.g., Castro-Larragoitia et al., 1997; Baig et al., 2010, 2011; Neidhardt et al., 2012; Drlíčková et al., 2013; Rosas-Castor et al., 2014a, 2014b), rice (e.g., Azizur Rahman et al., 2007; Bhattacharya et al., 2010; Khan et al., 2010; Hsu et al., 2012; Chou et al., 2014; Li et al., 2014; Liu et al., 2015), and wheat (e.g., Norra et al., 2005; Huang et al., 2008; Liu et al., 2009; Zhao et al., 2010; Baig et al., 2011; Bermudez et al., 2011; Álvarez-Ayuso et al., 2012; Tong et al., 2014). Conversely, other cereal crops such as rye (*Secale cereale* L.) have received relatively little attention (Schroeder et al., 2005; Mains et al., 2006; Gramss and Voigt, 2013). Although with much more reduced production, rye ranks as a world crop because of its wide cultivation area, including Russia, Belarus, Ukraine, Poland, Germany, Argentina, South Africa, Canada, and the United States (White et al., 2006; Wrigley and Bushuk, 2010). Rye is largely used as bread cereal, for alcoholic beverage manufacture, and for forage production. Other avenues comprise cover crop, green manure, and soil stabilization (Vinkx and Delcour, 1996; White et al., 2006; Ziolkowska and Jeleń, 2012). Rye has high-stress tolerance with respect to both soil and environment, being more competitive with other cereal crops on infertile, sandy, or acid soils (White et al., 2006; Newell and Butler, 2013). Rye has been also described as an As-tolerant species (Adriano, 1986). All these characteristics make rye a crop able to grow in highly degraded and As-polluted areas. The cultivation of rye under such circumstances may represent an important risk of As incorporation in the trophic chain with the consequent adverse effects on human health. In spite of all these aspects (wide cultivation area, different feed and food uses and ability to grow in As-affected areas), the knowledge about the As distribution in rye plants and its implications in the use of rye for feeding purposes is quite restricted.

The main objectives of this work are as follows: a) to perform the environmental characterization of agricultural soils impacted by the abandoned mine exploitation of an arsenical tungsten deposit, b) to evaluate the characteristics of As uptake, translocation and accumulation of rye plants cultivated in such soils, and c) to establish the suitability of rye edible parts for their safe consumption according to international legislation on food and feed.

2. Materials and methods

2.1. Study area

The studied mining area is situated 2 km southern of Barruecopardo village in the north-west of the Salamanca province (Spain) where the former Barruecopardo mine is located. This mine exploited extensively during the last century was the main tungsten deposit in Spain. Its metalliferous mineral association is formed by scheelite (CaWO_4), wolframite ($(\text{Fe,Mn})\text{WO}_4$), arsenopyrite (FeAsS), pyrite (FeS_2), chalcopyrite (CuFeS_2), and ilmenite (FeTiO_3), occurring in quartz veins hosted in granitic rocks. Mining activities generated great amounts of wastes composed of barren rocks, gangue minerals (mainly arsenopyrite), and their weathering products. Previous studies performed on this mining area such as screening of new plant species for phytoremediation have revealed high As levels in uncultivated soils (Otones et al., 2011a).

2.2. Sampling

Soil and plant collection was performed in a rye (*Secale cereale* L. cv. Petkus) cultivated field situated near a dump where mine tailings were accumulated (Fig. 1).

Soil sampling took place at 0, 25, 50, 75, 100, 125 and 150 m from the mine dump. Ten different samples were collected at each indicated sampling site. The soil profile was sampled in the depth up to 50 cm employing a 3-cm diameter core sampler. Samples corresponding to depths of 0–10, 10–20, 20–30, 30–40 and 40–50 cm were collected. Single composite samples were produced for the different soil depths at each of the mentioned sampling distances. After being air-dried, about 1-kg soil samples were sieved through a 2-mm mesh sieve for subsequent characterization.

Plant collection was also carried out at 0, 25, 50, 75, 100, 125 and 150 m from the mine dump. Sampling was carried out in the late spring (June). Rye plants near maturity with similar size were collected throughout the sampling area. Both roots and above-ground parts of the cultivated plant species were sampled. At least twenty plant specimens were sampled at the different collection sites. Rye plant samples were separated into roots, shoots, and grains. All plant sections were firstly rinsed with tap water, next cleaned employing an ultrasonic bath to remove soil dust particles, and finally washed with deionized water. Subsequently, cleaned plant samples were dried in an oven at 70 °C for 48 h, and then powdered for analysis.

2.3. Soil physico-chemical characterization

Soil samples collected at 0, 50, 100 and 150 m from the mine dump were characterized for their main physico-chemical properties. Such characterization was performed as follows: pH was measured potentiometrically using the saturated paste method, organic matter (OM) was obtained by dichromate oxidation using the Walkley–Black method (Walkley, 1947), the content of amorphous and poorly-crystalline Fe and Al oxides was determined by extraction with ammonium oxalate under darkness and the content of Fe and Al crystalline oxides by extraction with ammonium oxalate and ascorbic acid following the procedure described by Wenzel et al. (2001), available P was obtained by the Olsen sodium bicarbonate extraction method (Olsen et al., 1954) with P quantification by the spectrophotometric molybdenum blue method using

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