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Arsenic bioaccessibility in gold mine tailings of Delita, Cuba

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HIGHLIGHTS

- ▶ Tailings pile with up to 35,372 mg As kg⁻¹ are the main source of arsenic in Delita.
- ▶ Goethite and amorphous Fe arsenate are associated with maximum As bioaccessibility.
- ▶ The gastrointestinal As bioaccessibility is limited for arsenic in arsenopyrite and scorodite.

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ABSTRACT

A bioaccessibility test was carried out in four tailings collected at a former mining area in Delita, Cuba. A previous risk assessment study identified arsenic (As) as the main critical contaminant in this area and showed that the tailings had high As concentrations (up to 3.5%). This study aimed at: (i) evaluating As bioaccessibility in four tailings (R1, R2, R3 and R4) from a gold mining area to obtain a better health risk estimate; and, (ii) identifying the mineral phases responsible for most of the bioaccessible As using XRD, SEM-EDS, and XAS. The results showed that bioaccessible As in the tailings ranged from 0.65 to 40.5%. The main factors influencing As bioaccessibility were a high occurrence of amorphous iron arsenate; occurrence, even at low content, of iron oxyhydroxides and stability of mineral phases in the environment of the gastrointestinal tract. Although arsenopyrite, arsenates and goethite were confirmed by mineralogical methods such as optical microscopy, XRD, and SEM-EDS, XAS showed that scorodite–oxidation state As(+V)–was dominant in most of the tailings. This confirms that the low bioaccessibility of As in most of the tailings is due to the slow kinetics of As release from scorodite.

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1. Introduction

Arsenic (As) is a human health concern because it can contribute to skin, bladder, and other cancers [1]. The 51st most abundant element in crustal rocks (mean 1.8 mg As kg⁻¹), As occurs commonly in the environment in several oxidation states (–3, –1, 0, +3, and +5). In natural waters, it usually appears in the form of the oxy-anions arsenite [As(III)] or arsenate [As(V)] [2]. In general, reduced inorganic As(–III) found in sulfide minerals is relatively low in toxicity, but oxidized inorganic As(+III and +V) and its compounds are significantly more toxic than most of the organo-arsenic compounds. Moreover, As(+III) compounds are two to three times more toxic than As(+V) compounds [3].

Arsenic mobilization from rocks into the hydrosphere and pedosphere occurs in some areas with sulfide mineralization

(non-authigenic As source) and hence is a very common process in areas with volcanism or plutonism-related mineralization. Mineralized faults, epithermal stockwork and veins, massive sulfides, and hydrothermal conduits (which are often contained in altered rock that weathers more rapidly) are often produced by these conditions. The oxidation of authigenic As-bearing minerals is also important because it results in high concentrations of dissolved As in several aquifers around the world. Mining operations generally produce many types of wastes, including mine tailings, waste rock and slag. Specifically, mine tailings, which contain sulfide minerals, such as arsenopyrite (FeAsS), galena (PbS), and chalcopyrite (CuFeS₂) release As due to oxidative dissolution upon contact with the atmosphere and act as a principal source of environmental contamination in these areas [4].

To evaluate the actual As contamination in mining areas with sulfide deposits, three issues must be addressed: (a) the toxicity of the tailings, (b) the bioaccessibility of the elements and (c) environmental contamination analyses that identify the routes of exposure that place the population at the greatest risk [5].

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To evaluate toxicity, i.e. the ability under certain exposures to cause adverse effects on living organisms [1], different chemical extraction tests are commonly used [6]. Specifically, to assess the mobilization of contaminants from piles of tailings and media that are not exposed to organic acids (as assumed under the Toxicity Characteristic Leaching Procedure, TCLP), the gastric juice simulation test (GJST) is recommended [7,8]. However, some authors believe that this form of quantification does not take into consideration the toxicological interaction between different elements, and they suggest the use of *in vivo* tests to evaluate the total toxicity of the elements found in tailings [5]. Yet, *in vitro* tests are also a good choice for assessing arsenic health risks [9,10] and besides evaluating the gastric (like in GJST pH 2) and the gastrointestinal bioaccessibility (pH 5.5), such procedures could be used as a reasoning regarding the stability of the main mineral species potentially available in each compartment. In addition to pH and redox conditions, the composition of the mineral assembly [11] and the particle size [12] are important factors of As bioaccessibility, since these

factors are capable of controlling solubility of As-phases in mining tailings. The precipitation of Fe arsenates following the oxidation of arsenopyrite in general may limit As mobility.

The Delita gold mine region (Fig. 1), which is located in a watershed covering 72 km², has long been a matter of health concern due to possible surface and groundwater contamination with arsenic and heavy metals occurring in mine tailings and sediments [13]. In this area, a previous risk assessment study identified arsenic as the main critical contaminant [14,15]. The tailing piles located in the area correspond to remnants of the initial ore processing (grinding, separation, and calcination of the ore concentrate). In addition to the piles, arsenic and heavy metals have also been detected in weathered material from old mine tailings used to fill the path to a beach located close to the mine (Fig. 1). In the mine, tailings bearing arsenopyrite are exposed and As is transported by surface runoff toward the sea where arsenic was reported in concentrations up to 575 µg As L⁻¹ (Fig. 1). Moreover, water wells located in this area (V-28, S-7 e S-9) presented 11.6, 15.0 and 18.8 µg As L⁻¹ [14,15], which

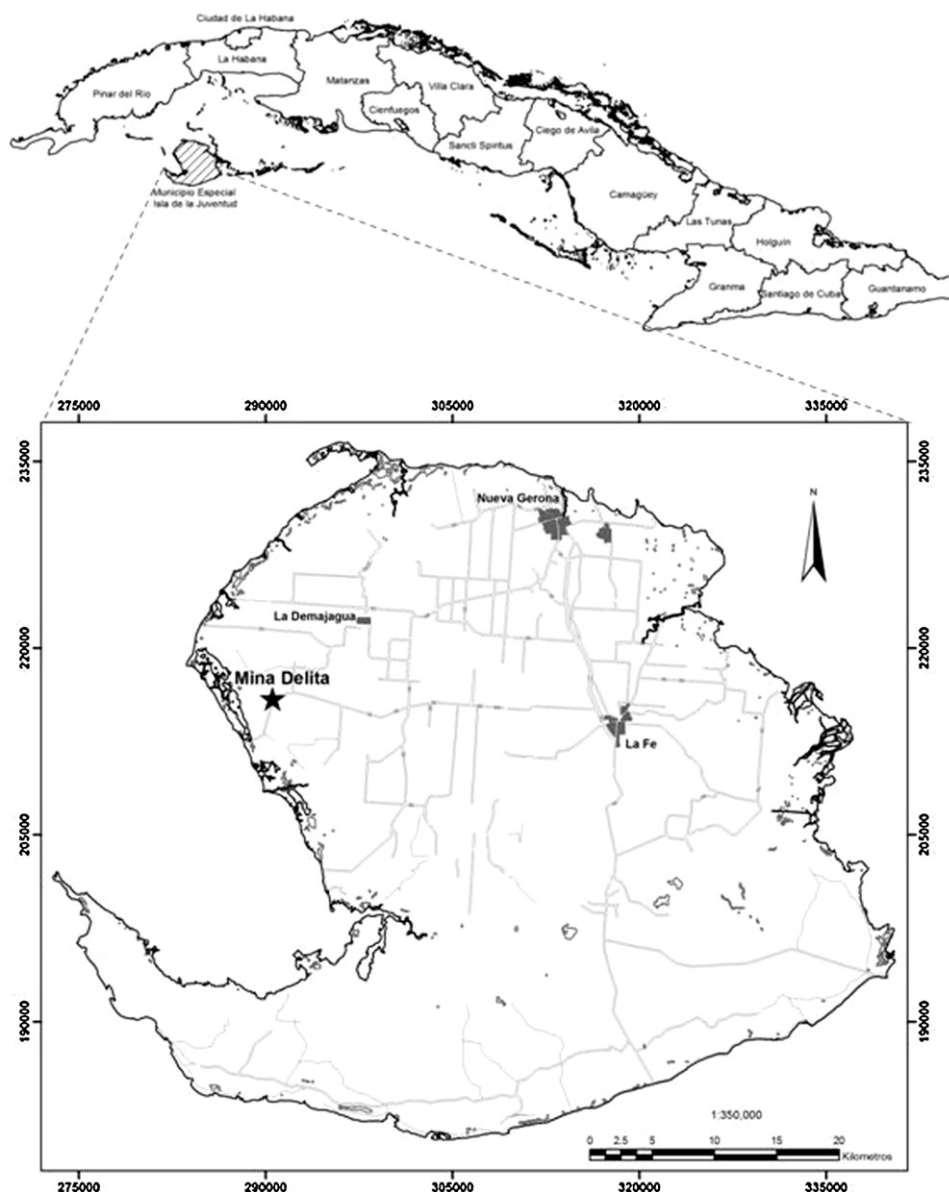


Fig. 1. Study area map.

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