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Processes controlling geogenic arsenic distribution in soils formed from iron-rich sedimentary rocks

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tent in soils formed from Fe-rich sedimentary rocks is

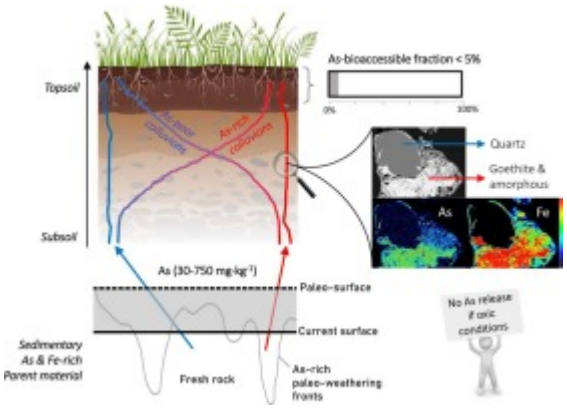
- Landscape/rock erosion contributes to spatial heterogeneity in soil parent material's As content.
- Exogenous processes partly explain geogenic As distribution in soils formed from Fe-rich rocks.
- Geogenic As is occluded in Fe oxyhydroxides in soils formed from Fe-rich sedimentary rocks.
- Geogenic As-bioaccessible fraction in topsoils formed from Fe-rich sedimentary rocks is ≤6%.

Abstract

Soils formed from Fe-rich sedimentary rocks can contain elevated As contents. Although the geogenic origin of As in these soils is recognized, the processes controlling its distribution within soil profiles and its mobility in topsoils are still unclear, limiting effective prediction of soils with potentially hazardous As contents for human health/ecosystems. We investigated 10 soil profiles (0–85 cm) formed from the As- and Fe-rich Aubange Formation in Belgian Lorraine. Sampling sites were chosen to represent the diversity of soil profiles associated with these rocks, with soil material collected every 5-cm for detailed As distribution assessment. Chemical and scanning electron microscopy analyses revealed that As is consistently associated with Fe oxyhydroxides variably enriched in K, Ca, Al and Si. X-ray diffraction identified goethite as a primary phase. The As-containing Fe oxyhydroxides originated from the soil parent material. They likely formed in response to fluctuating redox conditions during paleo-weathering of the Fe-rich sedimentary rocks and remained stable during pedogenesis. The As content varies widely within and across soil the profiles (23–753 $mg \cdot kg^{-1}$) and in topsoils (29–165 $mg \cdot kg^{-1}$), reflecting the chemical heterogeneity of the parent material. Additionally, exogenous processes, including colluvial transport and deposition of soil particles during and/or before pedogenesis, influenced As distribution within and across the soil profiles. Despite exceeding soil quality standards, As in the Fe-rich Aubange topsoils has

low bioaccessibility (3–6%) due to its strong association with the low-solubility Fe oxyhydroxides. Our findings emphasize the need for tailoring soil quality guidelines to the local pedo-geological context.

Graphical Abstract



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Introduction

Arsenic is a hazardous metalloid and a persistent pollutant that creates environmental, agricultural and health issues [2], [1]. A major exposure pathway for As is ingestion of soil particles or food grown in As-contaminated soil [3]. The ubiquitous presence of As in soil is linked to geogenic and/or anthropogenic sources [4]. Arsenic is a relatively abundant element in the Earth's crust (1.5–5.7 $mg \cdot kg^{-1}$; [5], [6]) and occurs in more than 300 different minerals, arsenopyrite (FeAsS) being the most common where reducing conditions prevail [7]. In oxidizing environments, As is often associated with ferric oxyhydroxides, such as goethite (α-FeO(OH); [8]). Potentially hazardous elevated As contents in soils may result from weathering of an As-rich parent material, typically sedimentary rocks [8]. Arsenic in topsoils may also increase due to anthropogenic inputs resulting from mining/smelting operations, burning of fossil fuels and biomass, use of pesticides and fertilizers in agriculture and industrial manufacturing (i.e. glass, ceramics, pigments, paints, etc.; [4]).

Many studies describe the occurrence and mobility of As in topsoils that have been affected by anthropogenic inputs and mining activities [9]. In contrast, topsoils with elevated As contents of geogenic origin have received less attention, despite concerns that the local population, particularly children, may be exposed to hazardous As levels through direct contact with the soil and/or via intake of locally grown food [10], [11]. Previous research on naturally occurring As in soils has been conducted primarily in relation to groundwater contamination [12] and its transfer to rice plants in paddy fields [13].

Recently documented occurrences of geogenic As in soils formed from As rich materials include variable contexts, such as soils formed from As-rich steatite (up to 225 $mg \cdot kg^{-1}$; [14]) and from arsenopyrite in gold sufide ores (> 30 $mg \cdot kg^{-1}$; [15]) in Brazil, or from unconsolidated materials (> 100 $mg \cdot kg^{-1}$; [16]) in Hong-Kong. Topsoils exhibiting high natural As contents, regularly exceeding 100 $mg \cdot kg^{-1}$ [17], [18], [19] are frequently associated with sedimentary rocks containing ferric oxyhydroxide-rich materials, such as “Iron Minette” (syn-sedimentary goethite/hematite ooids; [20]) and/or ferricretes (post-sedimentary iron-rich layer formed upon weathering in tropical climate [21]) are typically encountered in sedimentary basins, such as the Paris Basin [22]. These Fe-rich parent materials often contain a high As content, with values ranging from 435 up to 2900 $mg \cdot kg^{-1}$ [20], [23], [24], [25], [26]. They also tend to exhibit elevated contents of other potentially toxic elements, including Cr, Ni, Pb and Zn, which may be harmful to human health and ecosystems when present in soils.

The presence of geogenic As in topsoils poses a regulatory challenge, as their As content can approach or surpass established soil standards [18], [26], [27], [28]. While the geogenic origin of As in soils can be readily established, identifying potentially problematic soils in areas underlain by Fe-rich sedimentary rocks requires a detailed understanding of how parent material, pedogenesis and exogenic surface processes (such as erosion) influence the content and mobility of As in the topsoil. This entails to characterize the As-bearing solid phases existing in these soils and their evolution under the influence of soil-forming processes. Here we study As in soils formed from Fe-rich sedimentary rocks of the Aubange Formation, of Pliensbachian age, which is part of the Paris Basin in Belgian Lorraine. We analyze ten representative soil profiles collected in a 6-km^2 area as well as rock samples from outcrops or boreholes in the Aubange Formation. The studied soils have never been exposed to anthropogenic inputs of As [29]. Based on the soil stone mineralogy, soil geochemical signature and As-bearing phase composition, we shed light on the main pedogeological processes controlling the distribution of As in the soil profiles and across the study area. Additionally, we assess the bioaccessible fraction of geogenic As in the topsoils.

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Section snippets

Study area: geological background and rock sampling

The study took place across a 6-km^2 area in Belgian Lorraine (southern Belgium) in the commune of Aubange, which includes the village of Aix-sur-Cloie (Figure 1). Belgian Lorraine is located in the north of the Paris basin. The outcropping rocks belong to a Jurassic sedimentary sequence (Figure 1A), progressing from the youngest (Longwy Formation, Bajocian) in the south to the oldest (Ethe Formation, lower Pliensbachian) in the north. The study area is located in the Aubange Formation, which …

Soil profile descriptions

The soil texture is predominantly loamy, with a variable stone content (< 15–80 %), generally increasing with depth. Due to soil ploughing, the Ap horizon (topsoil: white line in Figure 3) ranges in depth from 0–18 to 0–34 cm in croplands (P1-P2-P4-P5-P6-P7-P8-P9) and from 0–10 to 0–18 cm in grasslands (P0-P3). In all but three soil profiles, the Ap-Bw-BC horizon sequence indicates a Cambisol. Profiles P2, P4 and P6 have a distinct Bt horizon and are classified as Luvisols. A minor increase in …

Total As content and geochemical signature

The soil As contents in profiles P0-P9 (Figures 1 and 4; Supp. Tables 4 and 5) always exceed the typical contents of European agricultural soils (< 3–20 mg·kg^{-1}, Q10-Q90; [46]) and are also higher than the worldwide average As content of pristine soils (6.8 mg·kg^{-1}; [47]). Excluding the P0 profile, the topsoil As content exceeds the Walloon standard value for agricultural soils (30 mg·kg^{-1}; [48]). High As contents of geogenic origin can occur in the surface horizons of soils developed on …

Conclusions

The ten agricultural soil profiles collected across a 6-km^2 area in Aubange reveal a wide range of As contents, often exceeding the Walloon soil quality standard. Notable variations in Mn and V contents, as well as in the potentially toxic elements Cr, Ni, Pb and Zn, were also observed. Pedogenesis took place in a geochemically heterogeneous soil parent material, originally comprised of calcareous sandstones and marls. This heterogeneity likely reflects multiple weathering episodes of the …

Environmental implications

The presence of As in the environment is a significant concern as this persistent metalloid poses potential toxicity risks to both human health and ecosystems. This study focuses on geogenic As in agricultural soils derived from iron-rich sedimentary rocks, where As levels often exceed regulatory thresholds, potentially posing a threat to human health and ecosystems. Our research identifies key processes controlling As distribution and mobility in these soils, highlighting the minimal …

CRediT authorship contribution statement

Van de Casteele Clélia: Investigation, Formal analysis, Conceptualization. **Philippe Sonnet:** Writing – review & editing, Formal analysis, Conceptualization. **Pierre Delmelle:** Writing – review & editing, Supervision. **Aubry Vandeuren:** Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Benoît Pereira:** Writing – review & editing, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. …

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. …

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