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Use of ecotoxicity test and ecoscores to improve the management of polluted soils: case of a secondary lead smelter plant

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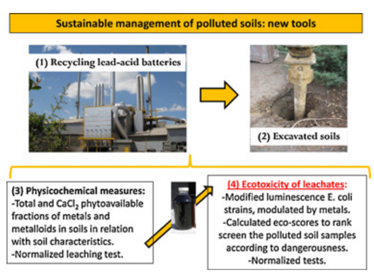
HIGHLIGHTS

- ► This article focuses on the use of biotests to improve polluted soil landfilling.
- ► New bioluminescent bacterial strains genetically modified were tested.
- ► Calculated ecoscores permit to rank polluted soil samples according to ecotoxicity.
- ► Results show the relevance of bioassays for polluted soils management.
- ► Total MTE-concentrations overestimate risks posed by contaminated soils.

GRAPHICAL ABSTRACT



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ABSTRACT

With the rise of sustainable development, rehabilitation of brownfield sites located in urban areas has become a major concern. Management of contaminated soils in relation with environmental and sanitary risk concerns is therefore a strong aim needing the development of both useful tools for risk assessment and sustainable remediation techniques. For soils polluted by metals and metalloids (MTE), the criteria for landfilling are currently not based on ecotoxicological tests but on total MTE concentrations and leaching tests. In this study, the ecotoxicity of leachates from MTE polluted soils sampled from an industrial site recycling lead-acid batteries were evaluated by using both modified *Escherichia coli* strains with luminescence modulated by metals and normalized *Daphnia magna* and *Alivibrio fischeri* bioassays. The results were clearly related to the type of microorganisms (crustacean, different strains of bacteria) whose sensitivity varied. Ecotoxicity was also different according to sample location on the site, total concentrations and physico-chemical properties of each soil. For comparison, standard leaching tests were also performed. Potentially phytoavailable fraction of MTE in soils and physico-chemical measures were finally performed in order to highlight the mechanisms. The results demonstrated that the use of a panel of microorganisms is suitable for hazard classification of polluted soils. In addition, calculated eco-scores permit to rank the polluted soils according to their potentially of dangerousness. Influence of soil and MTE characteristics on MTE mobility and ecotoxicity was also highlighted.

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1. Introduction

Originally located on the outskirts of cities, numerous industrial sites, sometimes abandoned, are now in urban areas and are therefore likely to have environmental and health risks to surrounding populations [1,2]. Currently, rehabilitation of the sites frequently entails excavation of polluted soils [3]. Excavated soils can thus follow two different ways: landfilling, expensive and energy intensive, or reuse/recycling, integrated to sustainable development. The choice of a specific track mainly depends on total and leachable concentrations of the pollutant in the soil [2]. Among the numerous pollutants observed in urban and peri-urban areas, trace metals are often present in soils [4]; atmosphere emissions by smelters being one of the main anthropogenic source [5,6]. MTE speciation and compartmentalization in soils can modify their impact on living organisms [5]. Now, numerous publications concluded that these two parameters are strongly influenced by soil organic matter (OM) content, pH and texture [7–9]. According to Matejczyk et al. [10], chemical weathering of soil minerals favours MTE solubilization and leachates production. Then, these leachates can pollute surrounding soils and waters. According to the council directive n°1999/31/CE, leaching tests with chemical analysis are therefore currently used for the assessment of environmental hazards of polluted soils. But, landfilling is often inevitable for strongly polluted soils, with high “hazard level” (assessed by leached and total MTE concentrations). Moreover, according to Foucault et al. [1], professionals consider the threshold set as too restrictive and they regret that excavated soils are almost always managed as waste.

In addition to the measure of total and leached MTE concentrations, it appears therefore that knowledge of MTE availability [11] and ecotoxicity may carry useful information [12–14] to improve environmental risk assessment [10]. Actually, the accurate estimation of metal phytoavailability in polluted soils and solid wastes, using single chemical extraction [15] carry interesting data to perform pertinent risk assessment and remediation efforts [16,17]. Soil quality integrates both physicochemical and biological characteristics [18]. Moreover, according to Plaza et al. [14] microorganisms play important roles in numerous soil functions. Soils are often polluted with a large variety of compounds leading to possible interactions [19], thus as reviewed by Kim and Owens [20] study of leachates ecotoxicity provides a direct functional characterization of various pollutant mixtures. But, only few studies concern the use of ecotoxicological tests to monitor contamination and bioremediation efficiency of polluted soils [21] and new tests are required by industrial sites managers to assess environmental risks. Among them, microbial bioassays offer quick, cheap and easy ecotoxicity (toxicity and mutagenicity) and bioavailability measurements on bacteria [22,23]. However, in many cases, microbial bioassays cannot be directly used for the identification and quantification of compounds due to the lack of specificity of the engineered microorganisms [24] and further studies are needed to improve these biotests.

The aim of this study was therefore to assess the ecotoxicity of leachates for landfilling of MTE contaminated soils by various complementary biotests, in addition to usual physicochemical measures. More precisely, the following two scientific objectives were aimed: (1) what is the pertinence of ecotoxicity tests to assess a more realistic human exposition to contaminated soil leachates? (2) What is the influence of soil physicochemical parameters on MTE mobility and leachates ecotoxicity? Several studies use specific bacteria strain to sense the presence of metals in soils [25–28], nevertheless, the development of statistical model to understand the link between chemical concentration of compound and bacteria sensors is still on-going work [23]. So, the originality of this study was to combine the use of new bacterial strains never tested in a context of the remediation of an industrial polluted site and calculation of eco-scores which facilitates the comparisons between different soils.

2. Materials and methods

2.1. Soil sampling and preparation

According to Wong et al. [29], the most relevant soil layer to study the environmental and sanitary impacts of MTE in urban areas is between 0 and 25 cm. Ten top soil samples (Fig. 1) were therefore collected in the courtyard of the Chemical Metal Treatments Society (STCM), a secondary lead smelter which currently recycles batteries located in the urban area of Toulouse (43°38′12″N, 01°25′34″E). This plant was chosen because of its activity and urban location, and many data are already available [1,4–6,30]. These data allowed defining different areas in terms of environmental and sanitary risks that can vary according to past and present activities. Moreover, previous studies of the particles released in the atmosphere by Uzu et al. [5] and Schreck et al. [4] revealed the presence of several MTE (Pb, As, Cu, Cd, Zn and Sb) and gave information on the main lead speciation: PbS, $PbSO_4$, PbO—$PbSO_4$, α-PbO and Pb (by order of abundance). All soil sampling points are presented in Fig. 1; they were dried, sieved under 2 mm and treated in triplicate.

2.2. Physico-chemical analysis

pH, organic matter and limestone contents, cation exchange capacity (CEC Metson) and texture, were determined for all soil samples, respectively, according to the norms ISO 10390 [31], ISO 10694 [32], ISO 10693 [33], NF X31-310 [34] and ISO 11277 [35]. Pb, As, Cu, Cd, Zn and Sb total concentrations were measured by ICP-OES (IRIS Intrepid II XXDL) after mineralization in *aqua regia* according to ISO 11466 [36] (HNO_3 65%, HCl 37%, ratio 1:3 (v/v)). The detection limits of Pb, Cd, Sb, As, Cu and Zn were 0.3, 0.2, 0.2, 0.2, 1.3 and 2.2 $\mu g\,L^{-1}$, respectively, whereas the limits of quantification were about 0.4, 0.3, 0.4, 0.3, 2 and 3 $\mu g\,L^{-1}$, respectively. The accuracy of measurements was checked using a certified reference material 141R (BCR, Brussels). The concentrations found were within 95–102% of the certified values for all measured elements.

2.3. Leaching tests

Normalized leaching test [37] was applied to all soil samples. This procedure consisted of a single extraction with deionised water, using a solid-to-liquid ratio of 1/10. 10 g of soil (granulometry at least < 4 mm according to the norm) was mixed with 100 mL deionised water during 24 h with end-over-end agitation at 5 rpm. After centrifugation at 3000 × *g* during 15 min, the leachates were filtered with cellulose 0.45 μm (Millipore®) filters. 10 mL of each leachates were then acidified with HNO_3 65% prior to analysis by ICP-OES (IRIS Intrepid II XXDL, analytical errors < 5%). The other part of leachates was not acidified so as not to disturb microorganisms used for further ecotoxicological tests.

2.4. MTE phytoavailability estimate

Potentially phytoavailable MTE concentrations were estimated by $CaCl_2$ extractions according to Uzu et al. [5]. In 25 mL polypropylene centrifugation tubes, 10 mL of 10^{-2} M $CaCl_2$ were added to 1.0 g of soil. The liquid to solid ratio of 10 is high enough to avoid samples heterogeneities [38]. After agitation end-over-end during 2 h at 5 rpm at 20 °C, samples were then centrifuged during 30 min at 10,000 × *g*. Supernatant was sieved through a 0.22 μm mesh and acidified at 2% with HNO_3 (15 N, suprapur 99.9%). MTE concentrations were finally measured by ICP-OES (IRIS Intrepid II XXDL,