



Contamination study of forest track soils located in a recreational area and filled with steel industry waste 30 years ago



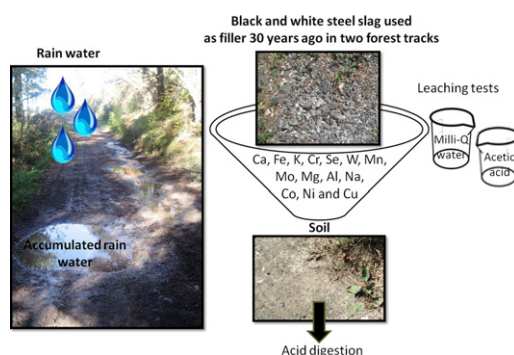
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HIGHLIGHTS

- Leaching of heavy metals from black and white steel slags used as filler in soils
- Milli-Q water and acetic acid leaching tests to simulate environmental conditions
- High levels of chromium in soils of forest tracks filled with steel slags
- Weathering of the steel slags caused by rain water

GRAPHICAL ABSTRACT



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ABSTRACT

The reuse of waste is increasingly widespread in order to avoid the exploitation of natural resources and to reduce costs. An example of that reuse is the employment of steel slag, a by-product from the steel making process. When the steel is produced through an electric arc furnace (EAF), two types of slag are generated: black and white slag. One application rarely used for this waste is as filler in forest tracks. In this work, two forest tracks of the Basque Country (northern Spain) filled with black and white slag 19 and 35 years ago, respectively, have been studied. Leaching tests were performed using Milli-Q water and acetic acid over the slags collected in that area. Additionally, soil samples collected near the slags were subjected to acid digestion. In these soil samples, there were elements of natural origin and others that could come from the leaching of the slag. Some of the more leached elements from the black slag (Ca, Fe, K, Cr, Se, W, Mn and Mo) and white slag (Mg, Al, Na, Co, Ni and Cu) coincided with the elements of highest concentration found in the soil samples. Moreover, there were differences in some elemental concentrations of soil samples with only black slag (higher presence of Ca and Mg) and soil samples with a mixture of both types of slag. It was noticeable that the highest concentration values of the measured elements were found on a specific side of the forest tracks, possibly due to the runoff water or the higher inclination of that side. On the other hand, some areas of both forest tracks could be considered contaminated by Cr according to a standard values from the Basque regulation, posing a risk to human health since they are recreational areas.

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1. Introduction

The activities related to the use and conservation of forest resources and the increasing demand of installations located in mountain areas

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(wind parks, antenna towers, telephone towers, etc.) require infrastructures to enhance the access to these places (Ansorena et al., 2003). Forest tracks are an example of such facilities. Depending on the topographical and climatologic conditions, the use of aggregates, concrete or slag can be essential so as to avoid the deterioration of the tracks and achieve their stabilization. The use of concrete for the construction of infrastructures is often unviable due to its high cost; therefore, steel slag is used instead, which is a by-product from steel production. The manufacturing of steel through an electric arc furnace (EAF) accounts for more than 40% of global steel production (Abu-Eishah et al., 2012), which in turn supposes a high generation of EAF steel slag. There are two types of steel slag, black slag and white slag, with different compositions according to their generation in the primary or secondary metallurgy (Yildirim and Prezzi, 2011). Moreover, their chemical components vary according to the furnace type, steel grades, pretreatment method, raw materials, etc. (Wang et al., 2010; Yi et al., 2012). Black steel slag has an expansive constitution because it contains lime oxides and free magnesium, which are easily hydrating and could provoke changes in its volume (Abu-Eishah et al., 2012; Pellegrino and Gaddo, 2009). On the other hand, the dissociation of $\text{Ca}(\text{OH})_2$, whose origin is in the hydration of CaO present in the slag, and the liberation of hydroxyl ions in environmental solutions would increase the solution pH. This could have a negative impact on ecosystems (Asaoka et al., 2013; Mayes et al., 2006; Mayes et al., 2008). White slag is chemically less complex and smaller than black slag (Radenović et al., 2013).

Generally, steel slag is thought to be one of the main contamination sources that affects the environment. This is attributable to the contamination by heavy metals in soil and nearby water where the slag has been accumulated. For example, high heavy metal concentrations due to the presence of slag have been found in mining areas (Manz and Castro, 1997), dump sites (Ganne et al., 2006) and in soil from the southern part of Madrid (Spain) where steel slag was deposited (García-Guinea et al., 2010). These situations could result in the accumulation of pollutants in vegetables and finally in the human food chain, posing a risk to human health. Moreover, wind erosion should be taken into account since it plays an important role in the spread of pollutants (Manz and Castro, 1997).

Despite the previous facts, EAF steel slag is classified as non-hazardous waste according to the European Waste Catalogue (European Waste Catalogue (EWC), 2000) and as inert industrial waste in the Basque Country legislation (BOPV, 1994). The Basque government regulates the construction of forest tracks and the use of steel slag in several applications by means of the 3/1998 law (General Law of Environment Protection of the Basque Country) (BOPV, 1998) and the 34/2003 decree about the treatment and subsequent use of the slag (BOPV, 2003). The later decree authorizes the use of black and white slag as a final product or raw material. However, it does not consider the possibility of mixing both types of slag for use. No legislation and no works have been found relating to this use. Nowadays, white slag is not used in construction (Prieto et al., 2011; Woltron et al., 2006), and mixtures of black and white steel slag are not employed as filling material due to the high economic value of the white slag (Chinhsiang et al., 2011). Nevertheless, the use of a slag mixture was found in past filling activities of forest tracks in the Basque Country. Considering the effects of metals in other forest soils (Chodak et al., 2013; De Vries et al., 2012), the main aim of this work was to analyze if the use of a slag mixture has harmful effects on the surrounding soil environment, at least in comparison with other forest tracks where only black slag materials were used. The study and analysis was performed on slag samples belonging to both types of forest tracks and in their adjacent soil samples. Leaching tests over the slags were conducted to determine the availability of the elements from the weathered slags to the surrounding soils. These tests were performed with Milli-Q water to reproduce rain water conditions over time and with acetic acid to simulate more extreme conditions, such as acid rain. Acid digestions over the soils were also carried out to ascertain whether or not the

metals were bound to the silicates. In addition, rain water was collected in the area of study. Finally, statistical analyses (correlation analysis and principal component analysis, PCA) were performed to analyze all of the concentration data (slag and soil) and to obtain additional relevant information from them.

2. Materials and methods

2.1. Materials

Samples were collected in two forest tracks located in Gipuzkoa (Basque Country, Spain) from 350 m above sea level to 200 m down, where they reach a secondary road. This area belongs to a temperate climate zone without drastic changes in weather. It has a very high regular rainfall (1051–1734 mm total annually), due to the latitude of the territory and its proximity to the Bay of Biscay as well as a mild temperature (11 °C on average annually) (The historic territory of Gipuzkoa, 2006). Furthermore, it has a rough topography with strong slopes. This area belongs to the Basin of the Urola River, but there are not aquifers. The forest track, with a mixture of slags, is located on a drainage area to a spring. Only runoff water could be considered to be the driving force to dissolve and transport heavy metals from the remaining slag to the surrounding soil.

The forest track with a mixture of slags has a length of 6 km and was built approximately 35 years ago (BWST). Additionally, a layer of white slag was added about 19 years ago in order to repair some sections. The other forest track is 0.5 km long and was constructed with only black slag 19 years ago (BkST). Both forest tracks are very close, but the BWST is located at a higher elevation and has more slope than the BkST (see Fig. 1.a Location of the forest tracks).

2.1.1. Long forest track (BWST)

Four slag samples, thirteen soil samples and two water samples were collected. Black and white slag samples were taken from the upper, middle and lower parts of the forest track. The same was done with the soil samples. Additionally, two more soil samples were gathered approximately 105 cm away from the forest track and at a 15 cm depth (where there were no remains of slag at the limit between the mixture of slag and soil and only soil). They were collected in order to observe possible differences with the rest of the soil samples. Furthermore, a farther sample was collected. It was considered to be a control sample because it was far enough (10 m) from the slag effect. Moreover, two water samples were collected from accumulated water on the forest track surface and directly from the atmosphere. Table 1 displays the descriptions of all of the samples.

2.1.2. Short forest track (BkST)

Four slag samples and five soil samples were collected due to the length of this forest track. As for the previous forest track, all of the samples were collected from the upper, middle and lower parts of the track. Table 2 shows the descriptions of these samples.

In both forest tracks, samples were collected taking into account the slope of the area and the runoff courses.

2.2. Methods

2.2.1. Pre-treatment

First, slag samples were ground by hand due to their large size and hardness, and afterwards, they were ground in a Fristch Pulverisette 6 mill (Indar-Orberstein, Germany) for 15 min with 2 repetitions of 700 rpm. These variables were altered according to the toughness of the slag. Second, they were lyophilized (Cryodos, Telstar) for 48 h to 150 mb and approximately $-52\text{ }^{\circ}\text{C}$. Finally, the ground and lyophilized samples were sieved. Grains less than 2 mm were used in the analytical procedure.

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