



## Lithological and land-use based assessment of heavy metal pollution in soils surrounding a cement plant in SW Europe



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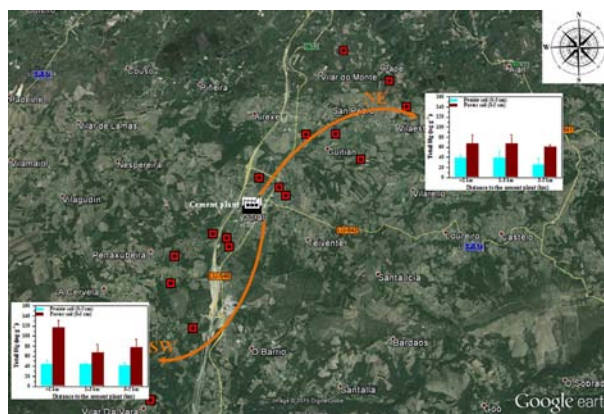
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### HIGHLIGHTS

- Land use influences Hg accumulated in soils nearby a cement plant in NW Spain.
- Total Cr and Pb contents in studied soils are similar to those in local lithology.
- Hg enrichment factor values in forest soils around the cement plant reach up to 36.
- Temporal flooding in prairie soils could favor the transformation of Hg to methyl-Hg.
- Cr mobility could be an issue of concern in the studied soils due to their acidity.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 14 December 2015

Received in revised form 28 March 2016

Accepted 29 March 2016

Available online xxxx

Editor: F.M. Tack

#### Keywords:

Mercury

Lead

Soil

Prairie

Forest

Enrichment factors

Soil pollution

### ABSTRACT

We study the influence of phasing out a cement plant on the heavy metal (Hg, Pb and Cr) content in the surrounding soils, taking into account factors often neglected, such as contributions due to local lithology or land use. The range of total Hg was 10–144  $\mu\text{g kg}^{-1}$ , reaching up to 41 and 145  $\text{mg kg}^{-1}$  for total contents of Pb and Cr, respectively. Forest soils showed higher concentration of Hg than prairie soils, indicating the importance of land use on the accumulation of volatile heavy metals in soils. In forest soils, total Hg showed a trend to decrease with soil depth, whereas in prairie soils the vertical pattern of heavy metal concentrations was quite homogeneous. In most cases, the distance to the cement plant was not a factor of influence in the soils content of the analyzed heavy metals. Total Pb and Cr contents in soils nearby the cement plant were quite similar to those found in the local lithology, resulting in enrichment factor values (EF's) below 2. This suggests that soil parent material is the main source of these heavy metals in the studied soils, while the contribution of the cement plant to Pb and Cr soil pollution was almost negligible. On the contrary, the soils surrounding the cement plant accumulate a significant amount of Hg, compared to the underlying lithology. This was especially noticeable in forest soils, where Hg EF achieved values up to 36. These results are of relevance, bearing in mind that Hg accumulation in soils may be an issue of environmental concern, particularly in prairie soils, where temporal flooding can favor Hg transformation to highly toxic methyl-Hg. In addition, the concurrence of acid soils and total-Cr concentrations in the range of those considered phytotoxic should be also stressed.

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## 1. Introduction

While weathering of parent material causes that heavy metals are naturally present in soils, industrial activities have resulted in a substantial increase in the pools of heavy metals in the pedosphere since the beginning of the XXth century (Han et al., 2002). The intensity of anthropogenic activities can disturb natural biogeochemical cycles of heavy metals in terrestrial ecosystems (Nriagu and Pacyna, 1998). Some anthropogenic activities considered as sources of heavy metals are fossil fuel combustion, ferrous and non-ferrous metal production, waste incineration and cement production (Pacyna et al., 2007, 2009).

The remarkable increase of population in urban areas taking place in the last decades has led to a large development in the cement industry world-wide. China, India and USA are the world leading countries in cement production (CEMBUREAU, 2015). In the European Union, Spain was the biggest producer until 2007, when the housing bubble begun to burst.

Cement facilities generate a great diversity of contaminants, mainly derived from energy consumption and use of raw materials. To obtain clinker, a cement precursor, raw materials (mainly limestone and clay) have to be calcined in rotary kilns, at temperatures around 1200–1500 °C, mainly achieved by means of fossil fuels combustion. Thus, fossil fuel combustion is considered one of the main sources of heavy metals release during cement manufacturing, in parallel to the occurrence of these metals in raw materials (Schuhmacher et al., 2002; Zemba et al., 2011; Rovira et al., 2014). Recently, used tires, sewage sludge, solvents or forestry residues have begun to be also used as alternative fuels, feeding kilns in cement facilities, acting as another source of heavy metals (Conesa et al., 2008, 2011; Schuhmacher et al., 2009; Zemba et al., 2011; Rovira et al., 2014). In addition, quarrying and grinding of the raw material, transportation, packing and dispatch of cement may result in the release of pollutants (Sikkema et al., 2011; Paoli et al., 2014).

Heavy metals present in the environment have been receiving increasing attention due to its potential negative effects on different ecosystem, and even on human health, mainly related to its potential toxicity, high persistence, non-degradation and bio-accumulation (Bozkurt et al., 2000).

The emission rate of heavy metals to the atmosphere from cement facilities is influenced by their different volatility during the manufacture process (Rovira et al., 2011a), as well as by the technology used to avoid their emissions from the stacks. Regarding heavy metal volatility, Conesa et al. (2008) distinguished among non-volatile (including Cr, Cu, Mn and Zn), semi-volatile (Pb, Cd) and volatile metals (Hg). After their emission to the atmosphere from the stacks of cement factories, heavy metals are finally transferred to the surrounding environment (soils, vegetation and water courses) through dry and wet deposition (Semhi et al., 2010; Rothenberg et al., 2010). The extent of the area of terrestrial ecosystems affected by heavy metal deposition from cement facilities depend on factors such as the type of metal emission (gas or particle), and meteorological parameters such as wind (direction, speed, frequency), precipitation, etc. Heavy metals released from cement facilities are reported to be accumulated in plants (Schuhmacher et al., 2002, 20; Isikli et al., 2003; Rovira et al., 2011b), lichens (Jóźwiak and Jóźwiak, 2009; Ljubic Mlakar et al., 2011; Paoli et al., 2014), and specially in soils (Asubiojo et al., 1991; Schuhmacher et al., 2004; Al-Khashman and Shawabkeh, 2006; Bermúdez et al., 2010; Rothenberg et al., 2010; Rovira et al., 2011a; Wang, 2013).

As soils are the most important sink of heavy metals in terrestrial ecosystems, this accumulation could lead to the loss of soil functions, such as those associated to environment quality and crop productivity. Potential toxicity to the biota is also frequently reported in heavy metal contaminated soils, with severity dependent on heavy metal bio-availability, which is related to soil characteristics such as pH, organic matter content, clay content and typology, cation exchange capacity, etc. Thus, the interactions between heavy metal and soils will finally

determine its long-term accumulation in soils, their possible transference to water courses or its potential entrance to the food chain through its accumulation in plants and crops grazed by livestock (Kabata-Pendias and Mukherjee, 2007).

In view of that, the aim of this study is to assess the content and distribution of heavy metals in soils at different depths and with a contrasting land use (forest and prairie soils), as well as considering different orientations and distances from a phasing-out cement facility. The study is focused on three heavy metals with different behavior in terms of volatility (Hg, Pb and Cr), considered of primary concern for human health and the natural environment. Moreover, Hg, Pb and Cr enrichment factors are estimated in the studied soils using their contents in the local lithology, a factor often neglected in similar studies, but needed to obtain a proper assessment of the influence of cement production on the accumulation of heavy metals in the surrounding soils. Finally, overall environmental implications will be also outlined.

## 2. Material and methods

### 2.1. Study area and soil sampling

The study area is located in Oural (42° 43'3.72"N, 7° 26'55.61" W), a small village in the south of the province of Lugo (NW Spain; Fig. 1) where a cement facility is operating since 1962. In times of plenty activity, the cement facility of Oural had a yearly production capacity of 450,000 tons of clinker, but since five years ago its activity is diminishing significantly. The facility is located in a rural landscape, far away from other industries and main highways, dominated by undulating terrain (elevation ranges between 450 and 750 m above sea level) covered by pastures and scattered small patches of deciduous forests, composed mainly by oak, with some chestnut and birch. The geology of the study area is dominated by different types of schists with similar petrology (micaceous schist or micaschists and slate schists) that belongs to the Villalba Series (Parga and Vegas, 1972), a Precambrian formation that cover territories located at the east and northeast of the cement plant where quartzites and gneiss are also present (Fig. S1, Supplementary Material). While at the west and northwest of the cement plant the geology is dominated by two-mica granites, several bands of metamorphic materials belonging to the Cándana Series consisting in quartzites, different types of slates, sandstones and a narrow band of amplites are scattered at the south of the study area. Finally, several patches of Tertiary sediments and Holocene floodplain sediments can be found disseminated throughout the territory. The climate is oceanic, with some continental features, showing a mean annual temperature of 11.2 °C, and mean rainfall 1010 mm year<sup>-1</sup>.

A total of 18 soil sampling sites were selected at three different fixed distances from the cement facility: <1 km, 1–3 km and 3–5 km. Nine soil sampling points were located at the NE of the facility, and the other nine sites at the SW of the plant, both in the wind directions prevailing throughout the year.

At each site, two contiguous prairie and deciduous forest stands were selected. In the pasture plots, three transects were delimited, and five points per transect (20 m of separation) were identified, where soil samples were collected at 0–5, 5–10 and 10–20 cm depth using a soil corer. Soil subsamples were pooled in a composite soil sample for each pasture stand and depth. In the deciduous forest stands, a representative area of the forest patch was selected, and a 1-m-long trench was dug in the soil down to 30 cm depth with a shovel. Samples of the mineral soil (mostly A horizons) were collected following the same depth intervals as in pasture soils. After sampling, all soil samples were double-bagged in plastic bags, stored at 4 °C and transported to the laboratory.

In order to obtain information about the background geochemistry of the zone, rock samples of different lithologies were collected from the bottom of each soil pit, bagged and transported to the laboratory. In the laboratory, rock samples were washed with abundant distilled

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