Geochemical anomalies of household dust in an industrialized city (Huelva, SW Spain)

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HIGHLIGHTS
• Sulphide like elements has been identified in household dust in the city of Huelva.
• Geochemical maps allowed to distinguish a zonation of its chemical composition.
• PCA confirmed a major industrial source (Cu-smelter) in household dust.

ABSTRACT
Geochanical anomalies of sulphide like elements (Cu, As and Cd) derived from the industrial activity have been identified in household dust of Huelva (SW Spain) using geochemical maps. Major and trace elements were analysed by ICP-OES and ICP-MS, respectively. Electron images of single particles were analysed by SEM-EDS in order to know their size, shape and composition. The geochemistry of the household dust has been compared to anomalies in deposition particles, PM10 and soils. A zonation has been observed: the eastern part of the city displays higher concentrations of sulphide like elements than the western part, supporting the origin of these elements related to the vicinity of industrial estates (Cu-smelter processes). Other domestic sources (e.g. wall painting) did not contain any geochemical anomalies related to sulphide like elements in household dust. Principal Component Analysis (PCA) was applied for grouping elements with similar sources, and reinforced the identification of a major industrial source in the eastern part. In this context, geochemical composition of household dust is considered as a fingerprint in order to identify industrial sources in the indoor air quality of Huelva.

Keywords:
Household dust
Major and trace elements
Geochemical map
Industrial estate

1. Introduction
Indoor air quality is a matter of growing interest in recent decades in urban environments. It has been estimated that adults spend close to a 90% of their lifetime indoors, including homes and workplaces exposed to household dust (US EPA, 1997). Dust can be re-suspended into the air in a high percentage (93%) and more than one-fifth of the dust may enter the respiratory system (Lu et al., 2014), representing a potential health risk depending on its chemical composition.

A main source that contributes to household dust is street dust, which originates from atmospheric particulate matter (PM) (Finalisson-Pitts...
The complex inorganic and organic chemical composition of PM has been referred as a "chemical cocktail" by some authors (Moreno et al., 2006). PM contains several toxic elements, including heavy metals and metalloids (e.g., Cd, Pb, As, Cu, and Cr) that are harmful for human health (Fontúrbel et al., 2011; Dockery and Pope, 1996; Nriagu, 1998; Zheng et al., 2013; Turner and Simmonds, 2006). Some worldwide studies show that street dust contributes 20–95% to household dust (Meyer et al., 1999; Rasmussen et al., 2001), being also an important source of metals that can get concentrated in household dust (Argyraki, 2014). Consequently, homes near industrial areas may have a higher metal concentration than dwellings far from industry (Rasmussen et al., 2013). Correspondingly, homes in the outskirts may have lower metal concentration than homes in the city centre, especially if they are surrounded by gardens or parks (Rasmussen et al., 2011, 2013). Some elements (e.g., Pb) have been found to have a significant seasonality in their concentrations (Petrosyan et al., 2006). Other relevant studies in Sydney show that Pb concentrations in household dust are not correlated with the improvement in the air quality (Chattoodhny et al., 2003).

In addition to street dust, other sources that contribute to the formation and the chemical composition of household dust are the effectiveness of ventilation system, the air exchange rates, house age, house material and roof type (Kim and Ferguson, 1993; Darus et al., 2012). There are also indoor sources (e.g., domestic combustion, cigarette smoke and wall paints) (Finalisson-Pitts and Pitts, 2000; Hassan, 2012; Lucas et al., 2014). Depending on the colour of the wall painting, certain metals accumulate in the household dust (e.g., Pb, Mn, Cd, Cu, Pb and Zn) (Tong and Lam, 2000). Additionally, heating practices represent a source of metals, especially for domestic coal combustion (Wang et al., 2013). However, metal concentrations are high in homes with electrical heating if they are not well ventilated (Rasmussen et al., 2001). According to the literature, indoor/outdoor Pb concentration ratios are >1 in residential areas with low or none impact from industry due to the indoor sources of Pb (Rasmussen et al., 2011).

There are few studies which consider the chemical composition of the household dust in cities affected by metallurgical activities (Fontúrbel et al., 2011). In this sense, the present work studies the geochemical composition of the household dust of Huelva (southwest Spain), a city surrounded by large industrial facilities with various types of activities (e.g., Cu smelter, phosphoric acid factory and petrol refinery), affecting its air quality (Fernández-Camacho et al., 2010; Sánchez de la Campa et al., 2011a). There are epidemiological studies that have reported a high cancer mortality rate in Huelva, related to environmental factors derived from industrial activity and social factors (Benach et al., 2003; López-Abente et al., 2006; Alguacil et al., 2014; Rodríguez-Barranco et al., 2014).

The objective of the present study is to identify the possible contribution of industrial sources to the household dust in the city of Huelva, characterizing geochemical maps by metals and metalloids concentrations. The geochemical composition of the household dust has been compared to the geochemical anomalies in deposition particles, PM10, and soil composition of the city. Principal Component Analysis (PCA) was applied for grouping elements with similar sources. In this context, chemical composition of household dust was evaluated as a possible fingerprint of historic register of air

Fig. 1. Study area, the windrose diagram (Environment Ministry of Spain, 2001) and division in two parts of the city according to predominant winds.

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