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Lead isotopes as tracers of anthropogenic pollution in urban topsoils of Mexico City

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ABSTRACT

A survey was performed to trace the main source of anthropogenic Pb pollution in Mexico City through Pb isotopic signatures ($^{208}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{206}Pb/^{207}Pb$, and $^{208}Pb/^{207}Pb$) from 103 urban topsoil (0–5 cm) samples. Those were collected in the metropolitan area of Mexico City and compared with isotopic compositions of leaded gasoline (LG), domestic Pb ores (DLO) and parent rock (PR). The isotope ratios (IRs) of Pb were determined by inductively coupled plasma quadrupole mass spectrometry (ICP-QMS) and total Pb concentration analyzed by wavelength dispersive X-ray fluorescence (WDXRF). The range of Pb concentrations levels in urban topsoil samples was 15–473 mg/kg. The IR values obtained for these samples were 37.965–39.718 ($^{208}Pb/^{204}Pb$), 18.375–19.204 ($^{206}Pb/^{204}Pb$), 1.177–1.218 ($^{206}Pb/^{207}Pb$) and 2.443–2.496 ($^{208}Pb/^{207}Pb$). Analyzed topsoil samples with low Pb content (<50 mg/kg) displayed high dispersion in $^{208}Pb/^{204}Pb$ values, which are determined by different natural sources. Samples with 51–200 mg/kg Pb content, shown low dispersion that revealed the mixing between the natural Pb and anthropogenic Pb. The assessment of the IR values shown that, as Pb concentration increases, a trend toward gasoline IR data has been observed. The results obtained by this research suggest that although the use of leaded petrol had been banned in Mexico since 1997, the Pb pollution in the urban topsoils due to the historical use of Pb in petrol is still significant.

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1. Introduction

The element Pb has four naturally occurring isotopes: ^{204}Pb (1.4%), ^{206}Pb (24.1%), ^{207}Pb (22.1%), ^{208}Pb (52.4%). ^{204}Pb is not radiogenic, whereas ^{206}Pb, ^{207}Pb and ^{208}Pb are produced by the radioactive decay of ^{235}U, ^{238}U and ^{232}Th. The isotopic study of Pb was initially used as a geochronologic tool, however a second practical application is to trace the source of Pb in environmental materials. The natural variations in Pb isotopic ratios (IRs) are useful to elucidate the origin of Pb, since the isotopic composition is characteristic for different sources. In general, Pb isotopic signatures of unpolluted soils are generally more radiogenic ($^{206}Pb/^{204}Pb = 18.5–19.5$) than polluted soils. This is due to that they are derived from weathered rocks in which the Pb IR has evolved with time, reflecting the initial U/Pb and Th/Pb content of the parent material (Hansmann and Köppel, 2000). In polluted soils, the polluting element has the isotopic signature of its source (singular or multiple). Regarding the abundances of the radiogenic isotopes (^{206}Pb, ^{207}Pb, ^{208}Pb) and the analytical instrumentation to measure them, the combination among them to get IR values, have proved to be an useful environmental tool to trace Pb sources. However, the normalization with regard to ^{204}Pb (non-radiogenic and less abundance isotope) provides a greater variability and information between reservoirs or sources (Komárek et al., 2008).

Owing to the design and manner in which mass analyses operate, this technique has been found to be limited in both, precision and accuracy (Townsend et al., 1998; Vanhaecke et al., 1998; Marguí et al., 2007).

During the last decade some factors that affect the accurate and precise isotope compositions analysis, such as instrumental mass bias phenomenon, have been carefully evaluated and discussed (Vanhaecke et al., 1998; Al-Ammar and Barnes, 2001). The mass bias phenomenon introduce serious errors to isotope ratio measured with ICP-QMS. A suitable mass bias correction procedure aims to improve accuracy and precision of the measurements. This correction involves the determination of the optimal dead time of the ion counting detection systems, mass bias evaluation using the Pb/Pb ratio of SRM 981 solution and the application of the mass

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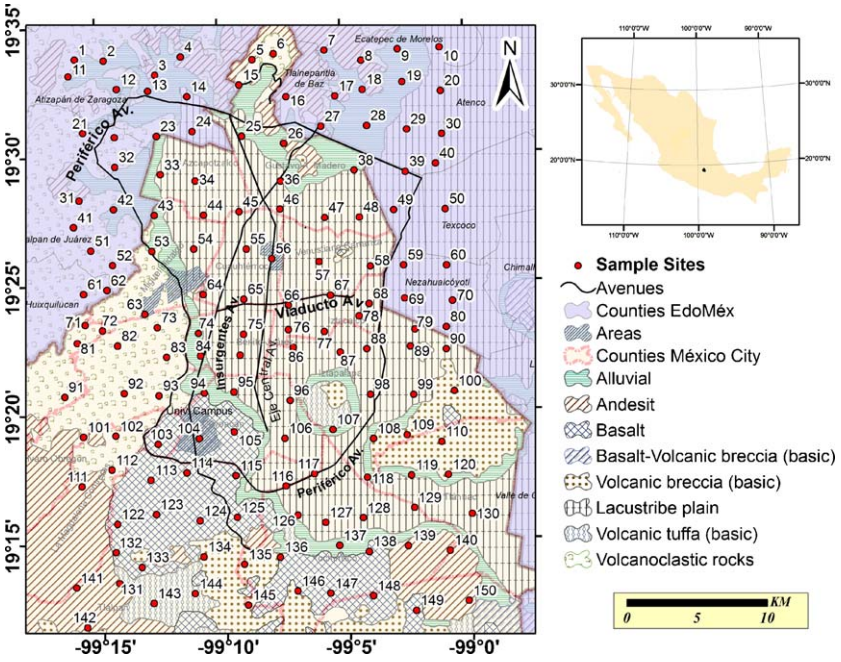

Fig. 1. Geological map of the metropolitan area of Mexico City and sampling points.

bias factor obtained to the reference material and/or real samples experimental data.

An internal standardization is employed to avoid instrumental drift. Thallium and Bi are used as internal standard allowing long data acquisition periods for isotope ratio measurement of Pb as analyte (Quétel et al., 1997; Adgate et al., 1998; Barton et al., 2000; Ettler et al., 2004; Marguí et al., 2007).

The degree of Pb contamination in urban environments has caused increasing concern during the last years. Even though many countries have restricted Pb as an additive in gasoline during the last two decades, it is widely recognized that combustion of leaded gasoline contributed to the major part of anthropogenic Pb found in the soils of urban environments (Murozumi et al., 1969; Shirahata et al., 1980; Graney et al., 1995; Watmough and Hutchinson, 2002; Wong et al., 2004; Komárek et al., 2008). The influences from other sources like industrial activities have also been reported (Mukai et al., 2001; Kurkjian et al., 2002; Chen et al., 2005). The differences in the nature and genetic origin of the analyzed soil samples must be considered when Pb isotope measurements are used for identification purposes. It is well known that soils from areas exposed to a rapid and uncontrolled urbanization are highly modified due to the intense human activities. Frequently in these cases, unpolluted soils with diverse genetic origin (and in consequence different Pb isotope ratio) are transported or/and relocated in the urban area. The isotopic composition of Pb in urban soils results from mixing many different sources, including natural and anthropogenic sources.

Early studies in the urban area of Mexico City have found elevated concentrations of heavy metals including Pb in urban topsoils, mainly related to vehicular traffic (Morton et al., 2001; Morton-Bermea et al., 2002, 2009; Rodríguez-Salazar et al., 2010b). In Mexico, Petróleos Mexicanos (PEMEX, 2004) reports in the chronology of the improvement of gasoline, that in 1986 the Tetraethyl lead content (TEL) changes from 3.5 to 0.5 ml/ gallon, while in 1994, the TEL content allowed was 0.1–0.2 ml/gallon, and in 1997 is removed the leaded petrol from the metropolitan area of Mexico City.

Soto-Jiménez and Flegal (2009) made an evaluation of different Pb pollution sources in the Gulf of California Ecoregion (northwest Mexico) and found a significant contribution from emissions produced by lead gasoline combustion used in the past. The Pb isotope ratios of environmental samples reported in this work were very similar to the average composition of Mexican Pb ores, based on the fact that Pb additives used in the Mexican leaded gasoline were predominantly derived from domestic Pb ores (Cumming et al., 1979).

The present research was performed in order to determine the natural Pb isotopic composition in the metropolitan area of Mexico City, determined mainly by the parental rocks, as well as the isotopic composition of possible anthropogenic sources and their influence, by measuring the Pb isotopic composition ($^{208}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{206}Pb/^{207}Pb$, and $^{208}Pb/^{207}Pb$) of 103 urban topsoils (0–5 cm) collected in the metropolitan area of Mexico City and the surrounding rural fields. The assess of this data will allow to establish the historical record of Pb contamination in the studied area. The isotope ratios were measured by ICP-QMS. The analytical methodology employed to analyze the IR values, was evaluated and reported by Rodríguez-Salazar et al. (2010a).

2. Materials and methods

The study area includes the metropolitan area of Mexico City as well as its suburban areas and surrounding rural fields and covers a total surface area of ca. 1200 km^2. Sampling points were chosen to cover gardens, parks, roadside fields, industrial sites and rural areas exposed to different pollution sources. A total of 103 topsoil samples (depth 0–5 cm) were collected during April–May 2008 employing rectangular grid sampling. Sampling locations were recorded by means of a GPS, and the data were geographically managed and processed with the GIS software ArcGIS (Environmental Systems Research Institute, Inc., ESRI, 2005), and Marco Geoestadístico Municipal (Instituto Nacional de Estadística, Geografía e Informática, México, 2005). Fig. 1 shows the distribution of sampling points in the study area.

As a parental rock, three replicates of the andesitic host rock surrounding the Valley (PR-1) were considerated. As possible anthropogenic sources, three replicates of domestic Pb ores from Naica Chihuahua (DLO-1) presumably used in the production of leaded gasoline in Mexico (Sañudo-Wilhelmy and Flegal, 1994; Soto-Jimenez et al., 2006; Soto-Jiménez and Flegal, 2009), were taken in account for analysis.

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