



**UNIVERSIDAD AUTÓNOMA DE GUERRERO**  
FACULTAD DE CIENCIAS QUÍMICO BIOLÓGICAS  
FACULTAD DE MEDICINA  
UNIDAD DE INVESTIGACIÓN ESPECIALIZADA EN MICROBIOLOGÍA

**MAESTRÍA EN CIENCIAS BIOMÉDICAS**

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**Distribución de fuentes de plomo en sangre de  
mujeres en edad reproductiva que viven cerca de  
jales en Taxco, Guerrero, México: un estudio  
isotópico**

**T E S I S**

QUE PARA OBTENER EL GRADO DE:  
**MAESTRA EN CIENCIAS BIOMÉDICAS**

**P R E S E N T A:**

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**Chilpancingo, Guerrero, Noviembre 2016.**

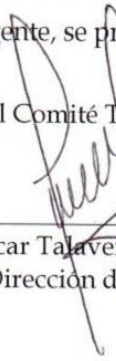


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APROBACIÓN DE TESIS


En la ciudad de Chilpancingo, Guerrero, siendo los 06 días del mes de julio de dos mil dieciséis, se reunieron los miembros del Comité Tutoral designado por la Academia de Posgrado de la Maestría en Ciencias Biomédicas, para examinar la tesis titulada **“Distribución de fuentes de plomo en sangre de mujeres en edad reproductiva que viven cerca de jales en Taxco, Guerrero, México: un estudio isotópico”**, presentada por la alumna Analine Berenice Vázquez Bahena, para obtener el Grado de Maestría en Ciencias Biomédicas. Después del análisis correspondiente, los miembros del comité manifiestan su aprobación de la tesis, autorizan la impresión final de la misma y aceptan que, cuando se satisfagan los requisitos señalados en el Reglamento General de Estudios de Posgrado e Investigación Vigente, se proceda a la presentación del examen de grado.

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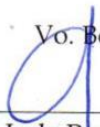
  
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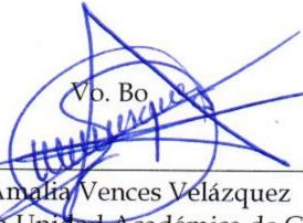
  
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Este trabajo se realizó en el Laboratorio de geoquímica de la Unidad Académica de Ciencias de la Tierra dependiente de la Universidad Autónoma de Guerrero y en el Laboratorio de Geoquímica Isotópica en el Departamento de Geociencias de la Universidad de Arizona.

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Durante el período en que cursó la Maestría en Ciencias Biomédicas, la Bióloga Analine Berenice Vázquez Bahéna, recibió beca del CONACYT con número (CVU): 555537.

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**Source apportionment of lead in the blood of women of reproductive age living near tailings in Taxco, Guerrero, Mexico: An isotopic study**

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## Title page

### Source apportionment of lead in the blood of women of reproductive age living near tailings in Taxco, Guerrero, Mexico: An isotopic study

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## **Abstract**

The concentration and isotopic composition of lead in the blood of women of reproductive age (15-45 y) exposed to multiple sources in two rural communities of the mining region of Taxco, Guerrero in southern Mexico were determined in order to identify specific contributing sources and their apportionment and to trace probable ingestion pathways. Our data indicate that more than 36 % of the studied women have blood lead concentrations above  $10 \mu\text{g dL}^{-1}$  and up to 87 % above  $5 \mu\text{g dL}^{-1}$ . Tailings contain between 2128 and  $5988 \text{ mg kg}^{-1}$  of lead and represent the most conspicuous source in the area. Lead contents in indoor dust are largely variable ( $21.7\text{-}987 \text{ mg kg}^{-1}$ ) but only 15 % of samples are above the Mexican Regulatory Guideline for urban soils ( $400 \text{ mg kg}^{-1}$ ). By contrast, 85 % of glazed containers (range:  $0.026\text{-}68.6 \text{ mg kg}^{-1}$ ) used for cooking and food storage are above the maximum  $2 \text{ mg L}^{-1}$  of soluble lead established in the Mexican Guideline. The isotopic composition indicates that lead in the blood of 40 % of the studied women can be modeled in terms of a simple binary mixing system between mineralization (and derivatives) and glazed pottery or mineralization and bedrock, end-members. In addition, another 45 % of the samples involve unleaded gasoline as a third source accounting together for 85 % of modeled samples. The remaining 15% of the samples involve a combination of these sources and mining leachates. Indoor dust is dominated by mineralization but some samples show evidence of contribution from a less radiogenic source very likely represented by interior paints but their influence in blood has not been recorded. This study supports the application of lead isotopic ratios to identify potential sources and their apportionment in humans exposed to multiple sources of lead from both, natural and anthropogenic origin.

**Key words:** Lead contamination; Natural/anthropogenic sources; Lead isotopic signature, binary/ternary mixing models; Southern Mexico.

## 1. Introduction

Lead is a highly toxic metal with no known biological function that produces serious adverse effects to biota and particularly to humans. It has been assessed that lead induces perturbations in the biosynthesis of hemoglobin producing anemia, atherosclerosis, cardiovascular disease, kidney damage and alterations of spermatogenic cells of men and of the hormonal profile in women decreasing fertility (e.g., Muntner et al., 2003; Hernández-Ochoa et al., 2005; Karita et al., 2005; Navas-Acien et al., 2007; Salih and Jaafar, 2013). Even though the weight of evidence is still weak, some studies have revealed that lead is directly or indirectly genotoxic and that the acute exposure to lead may induce lung, stomach and renal cancer (e.g., Healey et al., 2010). Based on epidemiological studies in humans and animals, the inorganic lead compounds are actually included by the IACR (2006) in the list of probable carcinogenic human substances. The effects of greatest concern, however, are those produced at the level of the central nervous system, which include serious perturbations in the hippocampus area and the synapses of neurons compromising the efficiency of memory, reducing the intelligence quotient, producing behavioral problems and diminishing school performance and learning, particularly in children, even at concentrations of blood lead as low as  $5 \mu\text{g dL}^{-1}$  (e.g., Lanphear et al., 2005; Garza et al., 2006; Rahman et al., 2011; Power et al., 2014). In chronically exposed pregnant women, lead may cross the placenta and then the hematoencephalic barrier of the fetus affecting its brain structure and the functionality of astrocytes and neurons, which in turn, diminish synapses and the liberation of neurotransmitters causing subsequent enduring neurobehavioral disorders, cognitive deficit and hyperactivity (Goyer, 1990; Goyer, 1996; Chuang et al., 2001; Surkan et al., 2008; Prins et al., 2010; Neal et al., 2011; Schneider et al., 2014). Lead exposure may continue during lactation because lead is significantly excreted via breast milk (e.g., Gulson et al., 1998; Ettinger et al., 2014) and thus, pregnant women and children are considered the most vulnerable population groups (Bellinger, 2008; Gulson, 2008; Moreno et al., 2010).

Exposure to lead is a serious public health problem worldwide and most countries have promulgated strict regulations to reduce exposure to the metal

particularly of children and women and have encouraged the development of effective analytical methodologies that allow the accurate quantification of lead in diverse biological fluids and tissues and the identification of potential point sources in both, indoor and occupational environments. Mines and smelters are the largest sources of emission of lead in the world but paints, batteries, tobacco and even cosmetics are also considered significant point sources of lead. In Mexico, glazed pottery is a major and widespread source of lead affecting a large portion of the population (Chaudhary et al., 2003) and leaded gasoline, although banned in México since 2000, its residues persist in soils and still represent a potential source of lead. The identification of potential lead sources and their distribution and availability are relevant issues in areas where there is founded evidence that the population is exposed to multiple sources in order to place constraints on the probable exposure pathways and to provide a suitable framework to undertake measures to reduce exposure to lead and associated toxic metals and to establish efficient remediation programs.

In this study, we measured the levels and the isotopic composition of lead in the blood of women of reproductive age exposed to multiple sources in two rural communities from the Taxco de Alarcón, Guerrero mining region in southern México. We performed the same measurements in the most potential sources such as mineralization, tailings, glazed pottery and indoor dust, which together with literature available and previously published isotopic data, allow us to establish an adequate data base to identify contribution of specific lead sources, their apportionment and to trace probable ingestion pathways.

## 2. Environmental framework

The mining region of Taxco de Alarcón is located in northern Guerrero State in southern México at 18° 23'-18° 48' N and 99° 30'-99° 47' W (Fig. 1). In the area, bedrock includes lower Cretaceous schist and metabasite of the Taxco Schist Formation, Albian-Cenomanian reefal limestone of the Morelos Formation, upper Cretaceous sandstone and shale of the Mexcala Formation, Paleogene red beds of the Balsas Formation and Oligocene acidic volcanic rocks of the Tilzapotla Formation (Campa and Ramírez, 1979). The Taxco mining district is famous worldwide for its more than 450 years of exploitation of base (Cu-Pb-Zn) and precious (Ag, Au) metals and during the 17-18<sup>th</sup> centuries, its mines were among the richest in America (e.g., des Riviers and Beals, 2011). Intensive ore exploitation resulted in the generation of large piles of solid wastes accumulated in the mine surroundings filling streams. Old wastes were buried or reworked and currently, only seven major tailings dams have been recorded in the Taxco area containing around 55 million tons of tailings (Fig. 1). The aerial disposal of tailings promoted their oxidation and the liberation of significant amounts of potentially toxic elements (PTE's) to the environment (Talavera et al., 2005; Romero et al., 2007). Affectation to surface water and stream sediments in the area has been documented by Talavera et al. (2006, 2016); Árcega-Cabrera et al. (2010); Méndez-Ramírez and Armienta-Hernández (2012) and Dótor et al. (2014). Furthermore, studies indicate that more than 10,000 ha of soil around the tailings have concentrations of PTE well above the regional background levels and that both, wild and crop vegetation shows severe impact by PTE (Gómez-Bernal et al., 2010; Ruiz-Huerta and Armienta-Hernández, 2012).

The El Fraile and La Concha tailings dams are located 5 km SW of Taxco City (Fig. 1). They were generated during the exploitation of individual mines in the period from 1940-1970 and contain about 5.5 and 2.5 million of tons, respectively of both, non-oxidized and highly oxidized tailings. Originally accumulated far away from the population, these deposits are now located within the rural communities of El Fraile and Santa Rosa, which together, have more than 2,500 habitants. The El Fraile and La Concha tailings are settled on carbonate-rich sandstone and shale of the Mexcala Formation (Fig. 1). Original, non-oxidized tailings are neutral (pH = 6.5-7.5) and

contain residual pyrite, chalcopyrite, galena, sphalerite, arsenopyrite and silver sulphides-sulfosalts in addition to several types of carbonates like calcite, manganoan calcite, rhodochrosite, siderite and kutnohorite (Talavera et al., 2005; des Riviers and Beals, 2011). Oxidized tailings are acidic (pH <6.5) and contain numerous secondary sulphates (gypsum, epsomite, hexahydrite, jarosite, pickeringite, bassanite, rozenite) and Fe-oxyhydroxides (hematite, magnetite, bernalite). According to Armienta et al. (2003); Talavera et al. (2005) and Romero et al. (2007), oxidized tailings of El Fraile and La Concha have the potential to release great quantities of PTE to the environment and available toxicological evidence indicates that children 6-11 years old in the zone contain anomalous levels of Pb in their blood and in addition, have As, Cr, Ni, Cd, Ba, Co, Cu, Zn, Mn, Mo, Sr and Fe in their urine (Moreno et al., 2010) and show evidence of DNA damage (Calderon-Segura et al., 2013).

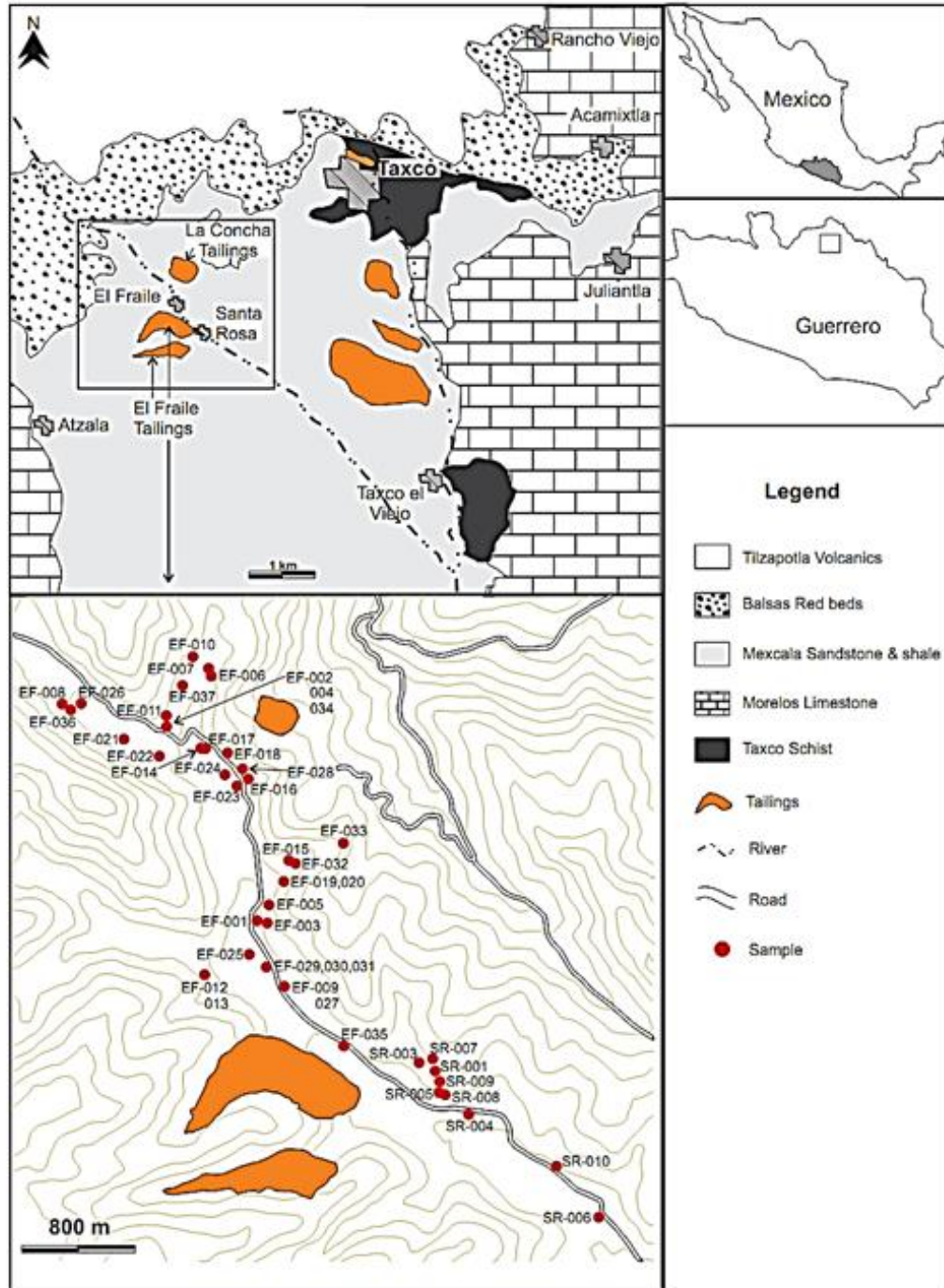


Figure 1. Geological map of Taxco, Guerrero (southern Mexico) showing the area of study and the location of the El Fraile and La Concha tailings dams. The location of houses where blood, indoor dust and/or glazed pottery utensils were sampled are shown in the inset map. Geology after Campa and Ramírez (1979).

### 3. Sampling and analytical methods

The concentration and isotopic composition of lead in the blood of forty seven women of reproductive age (15 to 45 y) were determined. The selected women are from the communities of El Fraile (n = 38) and Santa Rosa (n = 9; Fig. 1). All of the women who participated gave signed consent and only women living in the area for more than one year were included. Twenty seven samples of indoor dust from kitchens were collected and analyzed together with twenty six containers made of glazed pottery which are routinely used by women for cooking and food storage. Only in twenty cases were the three types of samples i.e. blood, dust and pottery obtained. Six primary minerals from the Taxco ore deposit and six representative samples from tailings (4 from El Fraile and 2 from La Concha) were also analyzed for lead concentration and isotopic composition.

Six mL of peripheral blood were taken by venipuncture and placed into vacutainer tubes with EDTA anticoagulant following the ethical code established in the declaration of Helsinki (WMA, 2013) and after written consent of the sampling procedure by the Bioethics Committee from the Autonomous University of Guerrero. Two aliquots of three mL each of blood were digested separately in ultraclean PTF Savillex bombs to determine lead concentration and isotopic composition, respectively. In both cases, blood was treated with repeated attacks of ultrapure  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$  on a hotplate following the procedure recommended by Gulson (2008).

Between 100 and 500 mg of ore minerals, tailings and indoor dust were accurately weighed in ultraclean PTF Savillex bombs and attacked with ultrapure 8M  $\text{HNO}_3$  and three times with inverted Aqua Regia on a hotplate at 120 °C to recover the environmentally available fraction. The sample was evaporated to near dryness and reconstituted with 1 mL of ultrapure 2%  $\text{HNO}_3$ . An aliquot of 0.5 mL was used for lead concentration and the remaining volume for isotopic determinations.

Containers of glazed pottery were washed three times with MQ water and then leached with 4% acetic acid at  $22 \pm 2$  °C during 24 hours according to the procedure established in the Mexican Regulatory Norm NOM-231-SSA1-2002. The volume lost during evaporation was restored by adding acetic acid to the initial volume and stirred



with a glass rod for 20 minutes. An aliquot of 6 mL was separated for lead concentration and the remaining volume was evaporated to near dryness and then redissolved with 2 mL of ultrapure 2% HNO<sub>3</sub> for isotopic determinations.

Lead concentrations were measured in triplicate in a Perkin Elmer Optima 3200 ICP-AES at the Laboratorio de Geoquímica, Universidad Autónoma de Guerrero. Accuracy and precision of analytical methods and instruments was monitored by using the certified standards INSPQ/Toxicologie-PC-B-L1108 for blood, CRM-LOAN-B for minerals, tailings and dust and, CWW-TM-A and CWW-TM-H for glazed pottery. Recovery of standards was 84-92% for blood, 91-97% for dust and 92-98% for glazed pottery. Based on the analysis of numerous multielemental standards over the last 20 years, the quantification limit for lead is better than 0.010 mg L<sup>-1</sup>.

The separation of Pb was performed by chromatographic methods using Sr Spect (Eichrom) exchange resin following the procedure outlined by Thibodeau et al. (2013). Lead isotope ratios were measured in ~50 ppb, Tl-spiked aliquots in an Isoprobe Multi-Collector Inductively Coupled Plasma Mass Spectrometer in the Laboratory of Isotopic Geochemistry of the Department of Geosciences, University of Arizona, USA. Isotopic fractionation was monitored by analyzing the NBS-981 standard with certified  $^{206}\text{Pb}/^{204}\text{Pb} = 16.9405$ ;  $^{207}\text{Pb}/^{204}\text{Pb} = 15.4963$  and  $^{208}\text{Pb}/^{204}\text{Pb} = 36.7219$  (Galer and Abouchami, 1998). The standard was analyzed once for every five unknowns. External errors associated with each Pb isotopic ratio are systematically < 0.05%.

## 4. Results

### 4.1. Lead concentrations

The concentrations of lead in analyzed samples are presented in tables S1 to S5 as supplementary material. The concentration of lead in the blood of women of reproductive age are globally variable ranging from 4.3 to 22.3  $\mu\text{g dL}^{-1}$  with an arithmetic mean  $\pm$  standard deviation of  $9.3 \pm 4.2 \mu\text{g dL}^{-1}$ . Beyond differences in the number of women studied in both communities, women from El Fraile have, on average, significantly higher lead concentrations ( $10.0 \pm 4.4 \mu\text{g dL}^{-1}$ ) relative to women from Santa Rosa ( $6.6 \pm 2.3 \mu\text{g dL}^{-1}$ ).

The lead content of primary minerals from the Taxco ore deposit is highly variable. Galena, the main lead ore, has 84.3%, which is near stoichiometric lead. Two samples of pyrite have concentrations of 966 and 1,055  $\text{mg kg}^{-1}$  whereas one sample of tennantite yielded 456  $\text{mg kg}^{-1}$ . Finally, a sample of rhodochrosite, a common gangue phase in the Taxco deposit, contains 357  $\text{mg kg}^{-1}$  of lead. The concentrations of lead in the tailings of El Fraile and La Concha range globally from 2128 to 5988  $\text{mg kg}^{-1}$  with an arithmetic mean  $\pm$  standard deviation of  $3638 \pm 1323 \text{mg kg}^{-1}$ . These values are within the range reported by Talavera et al. (2005) and no differences in the lead concentrations exist between the two deposits.

The concentrations of environmentally available lead in indoor dust are highly variable ranging from 21.7 to 987  $\text{mg kg}^{-1}$  with an arithmetic mean  $\pm$  standard deviation of  $230 \pm 226 \text{mg kg}^{-1}$ . Globally, dust from El Fraile has higher concentrations ( $287 \pm 238 \text{mg kg}^{-1}$ ) than dust from Santa Rosa ( $67.7 \pm 30.6 \text{mg kg}^{-1}$ ). The concentrations of leachable lead in glazed pottery are also variable ranging from 0.026 to 68.6  $\text{mg L}^{-1}$  with an arithmetic mean  $\pm$  standard deviation of  $14.7 \pm 18.7 \text{mg L}^{-1}$ . No significant differences exist between pottery from El Fraile ( $14.0 \pm 17.5 \text{mg L}^{-1}$ ) and Santa Rosa ( $16.8 \pm 23.3 \text{mg L}^{-1}$ ) as would be expected. Compared to the Mexican Guideline NOM-147-SEMARNAT/SSA1-2004, which establishes a maximum of 400  $\text{mg kg}^{-1}$  of total lead in urban soils, only ~15 % of samples (4 out of 28) are above this limit. Conversely, 85 % (22 out of 26) of glazed containers have lead concentrations higher than 2  $\text{mg L}^{-1}$ , the maximum limit established by the Mexican Regulatory Guideline for leachable lead in glazed pottery (NOM-231-SSA1-2002).

#### 4.2. *Lead isotopic composition*

The isotopic composition of lead in analyzed samples are presented in tables S1 to S5 as a supplement. Isotopic composition of lead in the blood of women of reproductive age from the two studied communities is rather variable with  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios varying from 18.6743 to 19.3931;  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios from 15.6398 to 15.7178 and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios from 38.4614 to 38.7994. The uranogenic  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios are in the range 0.8105-0.8375 whereas the thorogenic/uranogenic  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios are between 2.0007 and 2.0596. Although there is significant overlap, women from Santa Rosa have less radiogenic compositions compared to women from El Fraile.

Primary minerals of the Taxco ore deposit have a homogeneous lead isotopic composition with  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios of 18.7242 to 18.9812;  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios of 15.6513 to 15.8405 and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios of 38.7093 to 38.8047. The uranogenic  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios range from 0.8331 to 0.8359 whereas the thorogenic/uranogenic  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios are in the range of 2.0606-2.0676. This isotopic composition is indistinguishable from the historical isotopic signature reported by Cummings (1979) for Galenas from the Taxco ore deposit. On the other hand, tailings from the El Fraile and La Concha ponds also have a homogeneous isotopic composition with  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios of 18.7448 to 18.7527;  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios of 15.6519 to 15.6533 and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios of 38.7158 to 38.7190. The  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios ranges from 0.8347 to 0.8350 whereas the  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios are in the range of 2.0647-2.0654. This isotopic composition is within the range recorded in the ore minerals and is identical to the isotopic composition reported for metal-contaminated soils (Flores-Ronces, 2015) and tailings leachates (Talavera et al. 2016) in the zone.

Indoor dust showed the greatest lead isotopic composition with  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios of 16.9026 to 18.8784;  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios of 15.4783 to 15.6628 and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios of 36.6663 to 38.8047. The uranogenic  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios ranges from 0.8293 to 0.9153 whereas the thorogenic/uranogenic  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios are in the 2.0555 to 2.1693 range. Dust samples from Santa Rosa globally show a narrower isotopic composition falling, however, within the range recorded in the El Fraile samples (Table S4).

Glazed pottery also has a variable lead isotopic composition with  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios between 18.6850 and 19.2991;  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios of 15.6271 to 15.7054 and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios of 38.5044 to 38.8772. The uranogenic  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios ranges from 0.8134 to 0.8367 whereas the thorogenic/uranogenic  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios are in the 2.0145 to 2.0628 range. Glazed utensils from Santa Rosa have a narrower isotopic composition compared to those from El Fraile.

In addition to the isotopic composition of potential sources reported here, lead isotopes of other potential sources have previously been reported by the work group and by other authors including ore galenas from the Taxco ore deposit (Cummings, 1979); Mexican leaded and unleaded gasoline (Martinez et al., 2004; Morton et al., 2011); crop soils (Flores-Ronces, 2015); Taxco bedrock (except Morelos Formation which is reported here) and mining leachates (Talavera et al., 2016) and interior household paints (Rabinowitz, 1987), which are presented in tables S6 to S11 as supplementary material.

Table S1. Concentrations and isotopic composition of lead in the blood of women of reproductive age from the communities of El Fraile (EF) and Santa Rosa (SR) in the mining region of Taxco, Guerrero (southern Mexico).

Sample	Blood					
	Pb Conc $\mu\text{g dL}^{-1}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
EF-001	21.8	18.8586	15.6629	38.6725	0.83054390	2.0507
EF-002	8.1	19.1312	15.6916	38.6977	0.8202	2.0227
EF-003	11.0	18.8923	15.6668	38.6021	0.8293	2.0433
EF-004	7.6	19.0358	15.6802	38.6585	0.8237	2.0308
EF-005	22.3	18.8623	15.6670	38.6812	0.8306	2.0507
EF-006	11.0	18.9551	15.6679	38.6409	0.8266	2.0386
EF-007	14.4	18.8682	15.6671	38.6012	0.8303	2.0458
EF-008	10.4	18.9477	15.6776	38.5984	0.8274	2.0371
EF-009	7.5	18.9379	15.6647	38.6293	0.8272	2.0398
EF-010	8.6	19.0172	15.6772	38.6553	0.8244	2.0326
EF-011	5.3	19.1917	15.6920	38.7359	0.8176	2.0184
EF-012	14.3	18.8750	15.6619	38.6271	0.8298	2.0465
EF-013	13.7	18.8758	15.6661	38.6488	0.8300	2.0475
EF-014	7.1	19.3931	15.7178	38.7994	0.8105	2.0007
EF-015	6.7	19.0459	15.6765	38.7218	0.8231	2.0331
EF-016	12.6	18.8984	15.6682	38.6198	0.8291	2.0436
EF-017	10.0	19.1030	15.6882	38.7268	0.8212	2.0273
EF-018	8.6	18.9067	15.6650	38.6203	0.8285	2.0427
EF-019	19.7	18.8247	15.6628	38.7188	0.8320	2.0568
EF-020	7.5	18.9012	15.6609	38.6307	0.8286	2.0438
EF-021	11.3	18.9040	15.6701	38.6320	0.8289	2.0436
EF-022	15.6	18.7875	15.6506	38.6216	0.8330	2.0557
EF-023	11.8	18.8320	15.6583	38.5994	0.8315	2.0497
EF-024	8.4	18.8891	15.6632	38.6282	0.8292	2.0450
EF-025	11.1	18.8244	15.6455	38.6753	0.8311	2.0545
EF-026	8.1	18.9169	15.6759	38.6381	0.8287	2.0425
EF-027	6.4	18.8195	15.6571	38.5934	0.8320	2.0507
EF-028	9.1	18.8770	15.6629	38.6166	0.8297	2.0457
EF-029	9.7	18.8208	15.6428	38.6145	0.8311	2.0517
EF-030	9.0	18.8506	15.6539	38.6156	0.8304	2.0485
EF-031	6.6	18.8462	15.6563	38.6355	0.8307	2.0500
EF-032	4.8	18.8278	15.6529	38.6129	0.8314	2.0508
EF-033	5.4	18.8932	15.6639	38.6215	0.8291	2.0442
EF-034	6.2	18.8656	15.6646	38.5756	0.8303	2.0448
EF-035	5.1	18.8453	15.6569	38.6174	0.8308	2.0492
EF-036	10.1	18.8579	15.6624	38.5873	0.8306	2.0462
EF-037	6.0	18.8037	15.6550	38.5883	0.8325	2.0522
EF-038	5.9	18.7810	15.6510	38.5538	0.8333	2.0528

Table S1. (Continuation).

Sample	Blood					
	Pb Conc $\mu\text{g dL}^{-1}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
SR-001	10.1	18.7781	15.6485	38.6375	0.8333	2.0576
SR-003	6.8	18.8753	15.6638	38.5816	0.8299	2.0440
SR-004	7.6	18.8633	15.6545	38.5956	0.8299	2.0461
SR-005	6.2	18.8657	15.6621	38.6037	0.8302	2.0462
SR-006	4.9	18.8227	15.6558	38.5804	0.8318	2.0497
SR-007	9.4	19.0154	15.6778	38.6715	0.8245	2.0337
SR-008	4.8	18.8386	15.6533	38.6272	0.8309	2.0504
SR-009	4.3	18.8322	15.6580	38.5698	0.8314	2.0481
SR-010	4.7	18.6743	15.6398	38.4614	0.8375	2.0596
Min	4.3	18.6743	15.6398	38.4614	0.8305	2.0007
Max	22.3	19.3931	15.7178	38.7994	0.8375	2.0596
Mean	9.3					
SD	4.2					

Table S2. Lead concentrations and isotopic composition of selected primary minerals from the Taxco Ore Deposit.

Sample	Primary Ore Minerals					
	Pb Conc $\text{mg kg}^{-1}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
MP-4 Pyrite	1055	18.7242	15.6523	38.7139	0.8359	2.0676
MM-7 Sphalerite	832	18.7621	15.6532	38.7203	0.8343	2.0638
MR-8 Rhodochrosite	357	18.9812	15.8405	39.1813	0.8345	2.0642
MP-11 Pyrite	966	18.7858	15.6513	38.7093	0.8331	2.0606
MT-3 Tennantite	456	18.7534	15.6525	38.7195	0.8346	2.0647
MG-9 Galena	84.3%	18.7582	15.6524	38.7220	0.8344	2.0643
Min	357	18.7242	15.6513	38.7093	0.8331	2.0606
Max	84.3%	18.9812	15.8405	39.1813	0.8359	2.0676

Table S3. Lead concentrations and isotopic composition of representative samples from El Fraile and La Concha tailings impoundments.

Sample	Tailings					
	Pb Conc mg kg <sup>-1</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
El Fraile 02	2128	18.7527	15.6523	38.7186	0.8347	2.0647
El Fraile 06	4050	18.7518	15.6521	38.7162	0.8347	2.0647
El Fraile 11	5988	18.7448	15.6525	38.7158	0.8350	2.0654
El Fraile 19	3251	18.7511	15.6533	38.7184	0.8348	2.0649
La Concha 2	3568	18.7497	15.6519	38.7163	0.8348	2.0649
La Concha 9	2845	18.7500	15.6527	38.7190	0.8348	2.0650
Min	2128	18.7448	15.6519	38.7158	0.8347	2.0647
Max	5988	18.7527	15.6533	38.7190	0.8350	2.0654
Mean	3638					
SD	1323					

Table S4. Lead concentration and isotopic composition of indoor dust from the communities of El Fraile (EF) and Santa Rosa (SR) in the mining region of Taxco, Guerrero (southern Mexico).

Sample	Indoor Dust					
	Pb Conc mg kg <sup>-1</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
EF-001	240	18.4431	15.6218	38.3638	0.8470	2.0801
EF-002						
EF-003	280	17.8944	15.5703	37.7575	0.8701	2.1100
EF-004						
EF-005	166	17.5513	15.5383	37.3702	0.8853	2.1292
EF-006	579	18.7386	15.6506	38.7015	0.8352	2.0653
EF-007	612	18.7488	15.6504	38.7068	0.8347	2.0645
EF-008	164	18.7162	15.6509	38.7009	0.8362	2.0678
EF-009	478*	18.7739*	15.6532*	38.7254*	0.8338*	2.0627*
EF-010	324	18.7346	15.6519	38.6951	0.8355	2.0654
EF-011						
EF-012	139 <sup>†</sup>	18.7761 <sup>†</sup>	15.6538 <sup>†</sup>	38.6807 <sup>†</sup>	0.8337 <sup>†</sup>	2.0601 <sup>†</sup>
EF-013	139 <sup>†</sup>	18.7761 <sup>†</sup>	15.6538 <sup>†</sup>	38.6807 <sup>†</sup>	0.8337 <sup>†</sup>	2.0601 <sup>†</sup>
EF-014						
EF-015	392	18.7528	15.6533	38.7122	0.8347	2.0643
EF-016						
EF-017						

Table S4. (Continuation).

Sample	Indoor Dust					
	Pb Conc mg kg <sup>-1</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
EF-018	183	18.7460	15.6493	38.6876	0.8348	2.0638
EF-019	70.09‡	18.7941‡	15.6583‡	38.7398‡	0.8332‡	2.0613‡
EF-020	70.09‡	18.7941‡	15.6583‡	38.7398‡	0.8332‡	2.0613‡
EF-021	337	16.9026	15.4783	36.6663	0.9157	2.1693
EF-022						
EF-023	221	18.7373	15.6505	38.6946	0.8353	2.0651
EF-024						
EF-025	987	18.8784	15.6628	38.8047	0.8297	2.0555
EF-026	89.9	18.6999	15.6480	38.6791	0.8368	2.0684
EF-027	478*	18.7739*	15.6532*	38.7254*	0.8338*	2.0627*
EF-028						
EF-029	364▲	18.7473▲	15.6543▲	38.7107▲	0.8350▲	2.0649▲
EF-030	364▲	18.7473▲	15.6543▲	38.7107▲	0.8350▲	2.0649▲
EF-031	364▲	18.7473▲	15.6543▲	38.7107▲	0.8350▲	2.0649▲
EF-032						
EF-033	28.1	18.6955	15.6553	38.6696	0.8374	2.0684
EF-034		18.6099	15.6335	38.5327	0.8401	2.0706
EF-035						
EF-036	47.7	18.7206	15.6489	38.6845	0.8359	2.0664
EF-037	43.7	18.7029	15.6499	38.6783	0.8368	2.0680
EF-038						
SR-001	99.0	18.7880	15.6565	38.7252	0.8333	2.0612
SR-003	87.8	18.7260	15.6538	38.7167	0.8359	2.0675
SR-004	81.3	18.7243	15.6516	38.6927	0.8359	2.0664
SR-005	35.0	18.7951	15.6605	38.6387	0.8332	2.0558
SR-006	94.1	18.7422	15.6523	38.7090	0.8351	2.0653
SR-007	55.1	18.7133	15.6519	38.6936	0.8364	2.0677
SR-008						
SR-009						
SR-010	21.7	18.7446	15.6489	38.7059	0.8348	2.0649
Min	21.7	16.9026	15.4783	36.6663	0.8297	2.0555
Max	987	18.8784	15.6628	38.8047	0.9157	2.1693
Mean	230					
SD	226					

\* Women of samples EF-009 and EF-027 live in the same house and are exposed to the same dust.

† Women of samples EF-012 and EF-013 live in the same house and are exposed to the same dust

▲ Women of samples EF-029, EF-030 and EF-031 live in the same house and are exposed to the same dust.

‡ Women of samples EF-019 and EF-020 live in the same house and are exposed to the same dust.



Table S5. Lead concentration and isotopic composition of glazed pottery used by studied women for cooking and food storage.

Sample	Glazed Pottery					
	Pb Conc mg L <sup>-1</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
EF-001	0.026	18.8728	15.6656	38.6082	0.8301	2.0457
EF-002						
EF-003						
EF-004						
EF-005	16.27	18.7181	15.6383	38.6111	0.8355	2.0628
EF-006						
EF-007	4.04	19.2991	15.7054	38.8772	0.8138	2.0145
EF-008	3.16	18.9335	15.6785	38.5300	0.8281	2.0350
EF-009	3.63	18.7999	15.6461	38.6416	0.8322	2.0554
EF-010	0.678	18.6850	15.6414	38.5044	0.8371	2.0607
EF-011	68.6	18.7382	15.6345	38.6025	0.8344	2.0601
EF-012						
EF-013						
EF-014						
EF-015	5.67	18.8522	15.6654	38.7394	0.8310	2.0549
EF-016	13.0	18.8533	15.6680	38.5566	0.8310	2.0451
EF-017	2.82	18.7441	15.6271	38.6161	0.8337	2.0602
EF-018	44.3	19.0443	15.6904	38.6258	0.8239	2.0282
EF-019	0.127 <sup>‡</sup>	19.0304 <sup>‡</sup>	15.6929 <sup>‡</sup>	38.6320 <sup>‡</sup>	0.8246 <sup>‡</sup>	2.0300 <sup>‡</sup>
EF-020	0.127 <sup>‡</sup>	19.0304 <sup>‡</sup>	15.6929 <sup>‡</sup>	38.6320 <sup>‡</sup>	0.8246 <sup>‡</sup>	2.0300 <sup>‡</sup>
EF-021						
EF-022						
EF-023	5.36	18.7286	15.6338	38.6269	0.8348	2.0625
EF-024						
EF-025						
EF-026	17.6	18.8322	15.6646	38.5624	0.8318	2.0477
EF-027	16.7	18.7758	15.6435	38.6341	0.8332	2.0577
EF-028	24.1	18.7502	15.6403	38.6380	0.8341	2.0607
EF-029						
EF-030						
EF-031						
EF-032	25.8	18.9190	15.6691	38.6310	0.8282	2.0419
EF-033						
EF-034						
EF-035						
EF-036	1.71	18.9211	15.6736	38.5311	0.8284	2.0364
EF-037	12.2	19.0417	15.6975	38.6354	0.8244	2.0290
EF-038						

Table S5. (Continuation).

Sample	Glazed Pottery					
	Pb Conc mg L <sup>-1</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
SR-001	56.4	18.8546	15.6660	38.5834	0.8309	2.0464
SR-003						
SR-004	0.781	18.9391	15.6720	38.5686	0.8275	2.0365
SR-005	44.4	18.9407	15.6844	38.5705	0.8281	2.0364
SR-006						
SR-007	5.55	18.8703	15.6619	38.6418	0.8300	2.0478
SR-008	2.57	18.9084	15.6687	38.5978	0.8287	2.0413
SR-009	4.57	19.1462	15.6962	38.6813	0.8198	2.0203
SR-010	3.27	18.7640	15.6441	38.6125	0.8337	2.0578
Min	0.026	18.6850	15.6271	38.5044	0.8198	2.0145
Max	68.6	19.2991	15.7054	38.8772	0.8371	2.0628
Mean	14.7					
SD	18.7					

‡ Women of samples EF-019 and EF-020 live in the same house and use the same glazed pottery.

Table S6. Lead isotopic composition of galenas from the Taxco Ore Deposit reported by Cummings (1979).

Sample	Taxco ore galenas				
	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
TAX-5 Galena	18.748	15.658	38.747	0.8352	2.0667
786(J) Galena	18.739	15.643	38.691	0.8348	2.0647
787(SA-I) Galena	18.731	15.625	38.642	0.8342	2.0630
792(SA-EC) Galena	18.723	15.623	38.626	0.8344	2.0630
788(H) Galena	18.730	15.638	38.695	0.8349	2.0659
Min	18.732	15.623	38.626	0.8342	2.0630
Max	18.748	15.658	38.747	0.8352	2.0667

Table S7. Lead isotopic composition of tailings leachates from El Fraile impoundment used by inhabitants as source of domestic water. Data after Talavera et al. (2016).

Sample	Tailings leachates				
	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
<b>JF-01</b>	18.8523	15.6656	38.7255	0.8310	2.0542
<b>JF-02</b>	18.8553	15.6616	38.7216	0.8306	2.0536
<b>JF-03</b>	18.8592	15.6625	38.7200	0.8305	2.0531
<b>JF-04</b>	18.8523	15.6685	38.7211	0.8311	2.0539
<b>JF-06A</b>	18.7528	15.6536	38.7209	0.8347	2.0648
<b>JF-06B</b>	18.7520	15.6550	38.7190	0.8348	2.0648
<b>JF-07</b>	18.7514	15.6517	38.7124	0.8347	2.0645
<b>JF-08</b>	18.7536	15.6527	38.7160	0.8347	2.0645
<b>Min</b>	18.7514	15.6517	38.7124	0.8305	2.0531
<b>Max</b>	18.8592	15.6685	38.7255	0.8348	2.0648

Table S8. Lead isotopic composition of soils from Santa Rosa. Data after Flores-Ronces (2015).

Sample	Soils				
	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
<b>STR 2</b>	18.7422	15.6481	38.6993	0.8349	2.0648
<b>STR 10</b>	18.7506	15.6530	38.7119	0.8348	2.0646
<b>STR 11</b>	18.7539	15.6553	38.7192	0.8348	2.0646
<b>STR 14</b>	18.7094	15.6544	38.6426	0.8367	2.0654
<b>Min</b>	18.7094	15.6481	38.6426	0.8348	2.0646
<b>Max</b>	18.7539	15.6553	38.7192	0.8367	2.0654

Table S9. Lead isotopic composition of bedrocks from the Taxco mining area. The lead isotopic composition of the Morelos Formation is from this work; the remaining are from Talavera et al. (2016).

Formation	Bedrocks				
	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
Taxco Schist	19.7800	15.7042	38.9708	0.7939	1.9702
Morelos	33.3219	16.3309	38.5824	0.4901	1.1579
Mexcala	18.8105	15.6397	38.6710	0.8314	2.0558
Balsas	18.9640	15.6737	38.7850	0.8265	2.0452
Tilzapotla	18.7256	15.5933	38.4978	0.8327	2.0559
Min	18.7256	15.5933	38.4978	0.7939	1.9702
Max	19.7800	15.7042	38.9708	0.8327	2.0558

Table S10. Lead isotopic composition of actual unleaded and historical leaded gasoline from Mexico. Data after Martínez et al. (2004) and Morton et al. (2011).

Sample	Mexican gasoline				
	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
Unleaded MAGNA	18.0370	15.7610	36.8840	0.8740	2.0449
Unleaded PREMIUM	17.9380	15.7970	36.9960	0.8810	2.0624
Unleaded PREMIUM UG	18.2730	15.4203	37.5070	0.8439	2.0526
Leaded gasoline LG-1	18.7310	15.5573	38.3760	0.8306	2.0488
Leaded gasoline LG-2	18.6890	15.5483	38.4000	0.8319	2.0547
Min	17.9380	15.4203	36.8840	0.8306	2.0449
Max	18.7310	15.7970	38.4000	0.8810	2.0624

Table S11. Range of lead isotopic composition reported by Rabinowitz (1987) for interior paints from United States.

Sample	USA Interior Paints				
	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
Paints	17.5			0.84	
	19.4			0.91	

## 5. Discussion

### 5.1. Relationship of concentrations of lead in blood with major potential sources

Women of reproductive age from the rural communities of El Fraile and Santa Rosa in the mining region of Taxco, Guerrero (southern Mexico) have moderate to high concentrations of lead in their blood. Nearly 36 % (17 out 47) of the studied women have concentrations above  $10 \mu\text{g dL}^{-1}$  and 87% (42 out 47) above  $5 \mu\text{g dL}^{-1}$ , which are considered as levels of reference above which there are proven toxicological effects (CDC, 2012). According to the maps of figures S1 to S3 presented as supplements, women with levels of lead higher than  $10 \mu\text{g dL}^{-1}$  live downslope north of the El Fraile and La Concha tailings deposits (Fig. S1), a distribution strikingly comparable to that observed in the indoor dust samples with the highest concentrations of lead (Fig. S2) but very different from the distribution of concentrations of lead in glazed pottery (Fig. S3). These facts strongly suggest that: (1) lead from tailings is a significant contributor to the lead of indoor dust, a conclusion supported by lead isotopes as substantiated later; (2) indoor dust is an important source of lead in blood in the studied women and other household residents; (3) there is no correlation between the lead concentration of blood of the women nor in indoor dust with the distance of tailings. Studied women and dust samples from Santa Rosa, though closer to the El Fraile tailings, have lower lead concentrations than many women and dust samples farther north of the tailings deposit; (4) wind direction and topography are relevant factors controlling the direction of dispersion of the particles from tailings; and, (5) lead in glazed pottery seems to play, at first sight, a second-order role in the concentration of lead in the blood of women, however, this is not accurate because as evidenced by the isotopic composition (see below), glazed pottery is a major contributor to the lead in their blood.

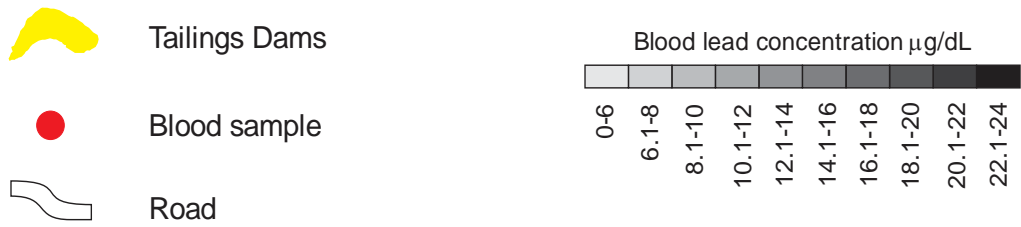
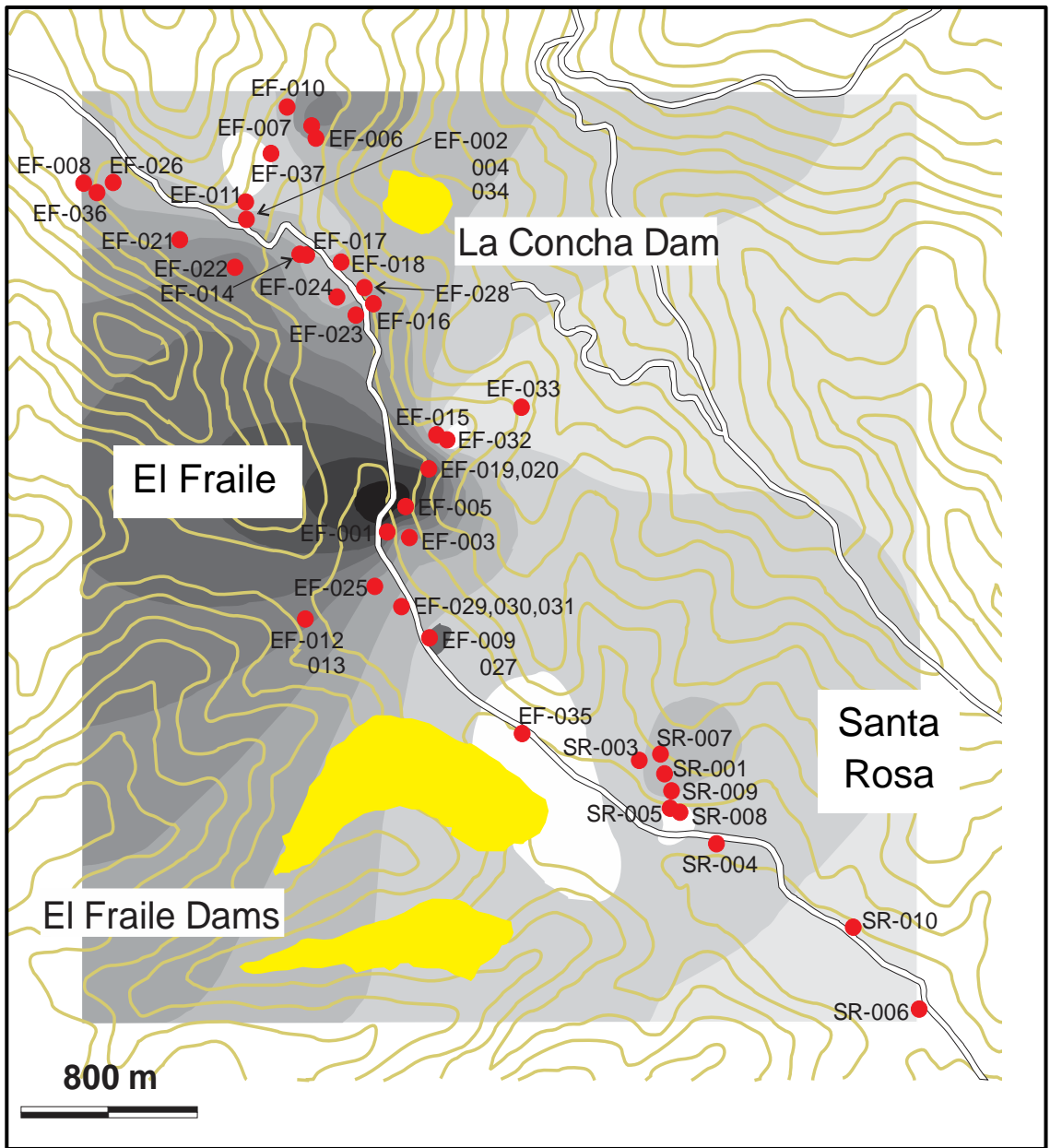


Figure S1. Map of isoconcentrations of lead in the blood of women of reproductive age from the communities of El Fraile and Santa Rosa in the mining region of Taxco, Guerrero in southern Mexico.

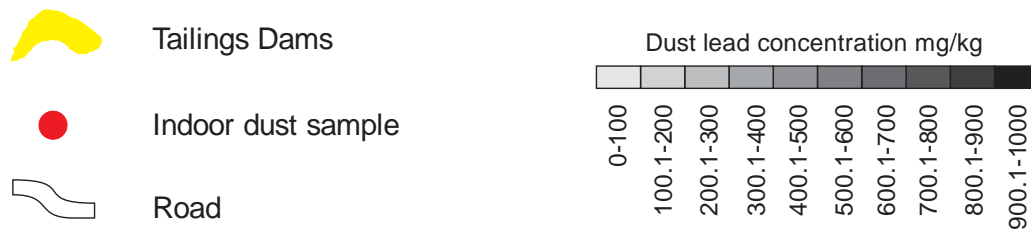
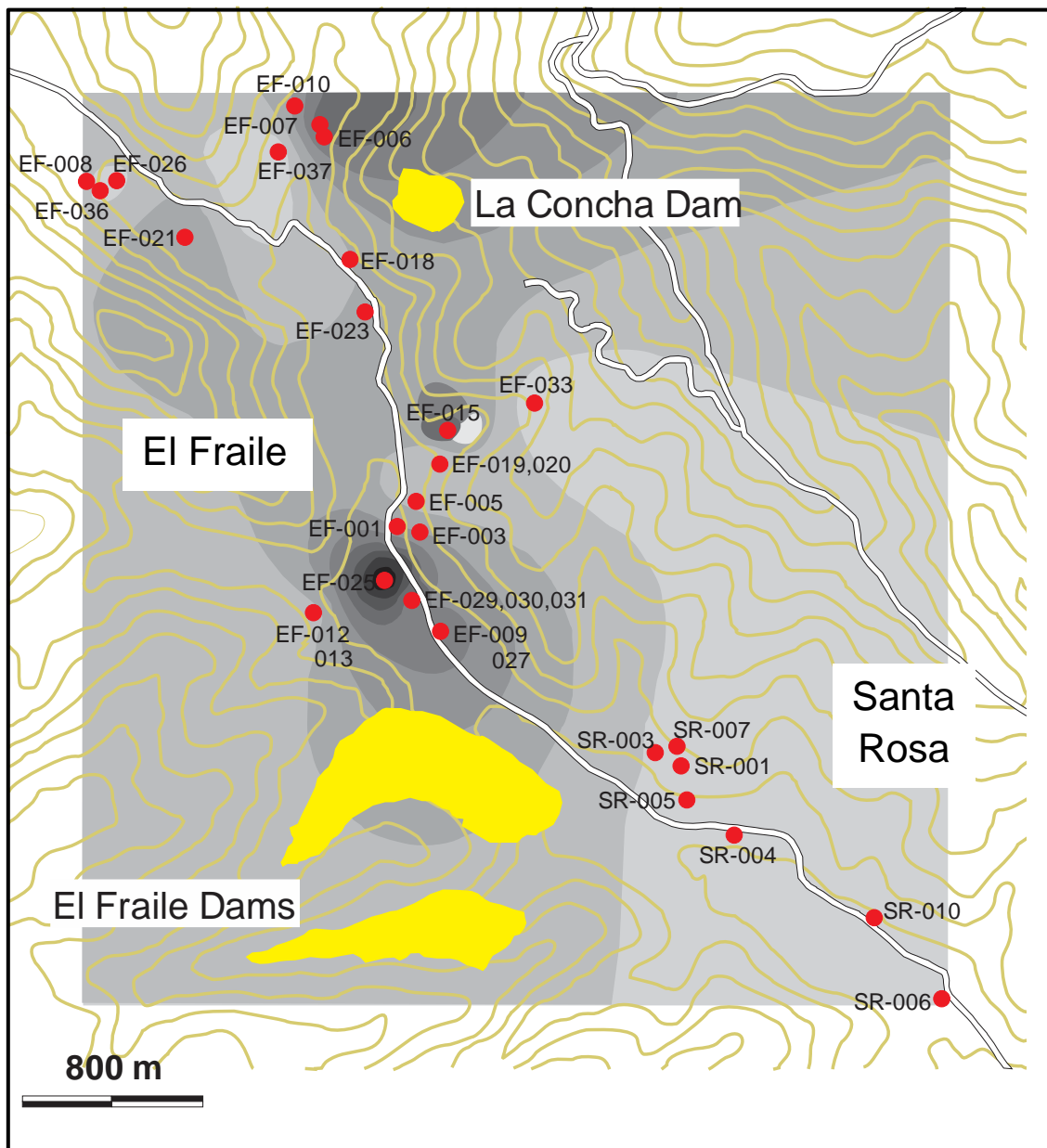


Figure S2. Map of isoconcentrations of lead in dust of houses from the communities of El Fraile and Santa Rosa in the mining region of Taxco, Guerrero in southern Mexico.

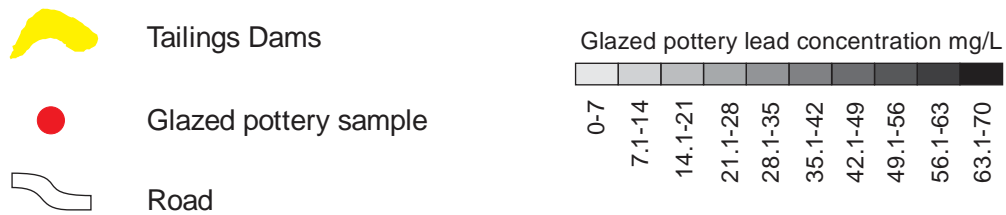
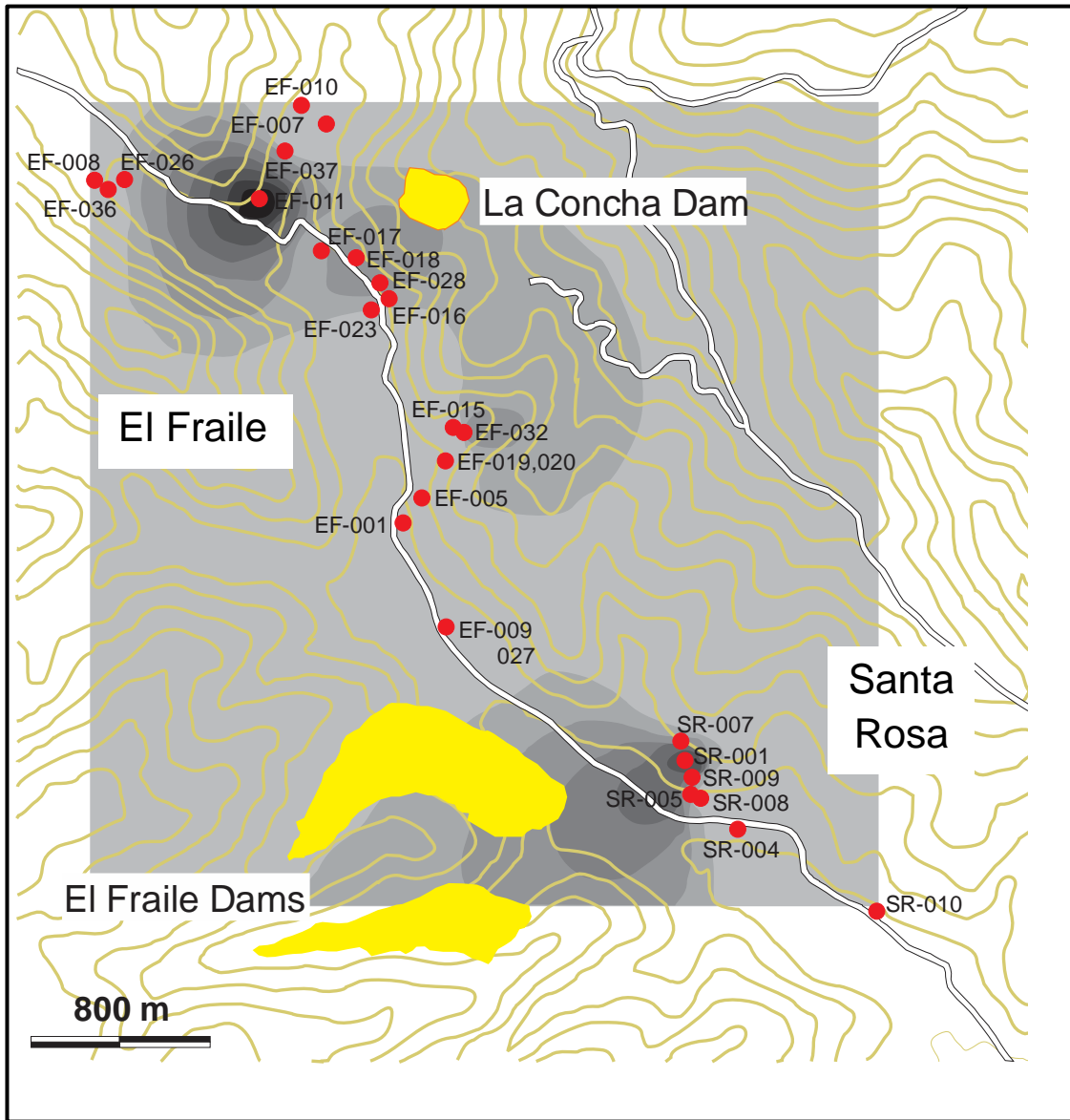


Figure S3. Map of isoconcentrations of lead in glazed pottery used by women from El Fraile and Santa Rosa for cooking and food storage.



## 5.2. Source identification and apportionment

Lead isotopes undergo no mass-dependant fractionation by any known physical, chemical or biological process and therefore, they constitute a powerful tool for identifying point sources of lead and associated metals in polluted environments and utensils (e.g., Rabinowitz, 1987; Chaudary et al. 2003; Faure and Mensing, 2005; Kamenov and Gulson, 2014). The isotopic fingerprint of lead in the sources and contaminated materials is commonly defined by the three major ( $^{206}\text{Pb}/^{204}\text{Pb}$ ;  $^{207}\text{Pb}/^{204}\text{Pb}$ ;  $^{208}\text{Pb}/^{204}\text{Pb}$ ) isotopic ratios, which are frequently complemented by other combinations such as the  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios or their reciprocals. It is assumed that if one object with a particular lead isotopic composition matches within analytical error the isotopic composition of a specific source in a given area, then, it is almost certain that lead in the object derives from that source (e.g., Rabinowitz, 1987; Gulson, 2008). In many cases, however, matching is not perfect in all the isotopic ratios due to contribution from more than one lead source and thus, mixing vectors must be established to identify potential sources and their apportioning.

In the area, several potential point sources of lead are evident including ore deposits, tailings, soils, leachates, indoor dust, glazed pottery, bedrock, gasoline and interior paints. Figure 2 shows the range of the principal ( $^{206}\text{Pb}/^{204}\text{Pb}$ ;  $^{207}\text{Pb}/^{204}\text{Pb}$ ;  $^{208}\text{Pb}/^{204}\text{Pb}$ ) and complementary ( $^{207}\text{Pb}/^{206}\text{Pb}$ ;  $^{208}\text{Pb}/^{206}\text{Pb}$ ) isotopic ratios of lead in the blood of the studied women together with the isotopic range of available potential sources in the area. From this figure, it is evident that the isotopic composition of blood does not match fully with any individual source in all isotopic ratios indicating that lead in the blood most probably derives from multiple sources. It is, however, also evident that the isotopic composition of glazed pottery and mineralization (and its derivatives like tailings, soils and leachates) is globally very similar to the isotopic composition of blood and strongly suggests that both, pottery and mineralization may represent significant contributors to lead in the blood of the women in the studied area. Most samples of indoor dust have the same isotopic composition as mineralization and its derivatives and blood strongly suggesting that dust may also represent an important pathway of lead. Matching of the isotopic composition of blood

with other potential sources is partial or lacking which might imply that they are minor contributors (or not at all) to lead in the blood of the studied women.

Correlation of uranogenic ( $^{207}\text{Pb}/^{206}\text{Pb}$ ) and thorogenic/uranogenic ( $^{208}\text{Pb}/^{206}\text{Pb}$  or  $^{208}\text{Pb}/^{207}\text{Pb}$ ) ratios have often been used to show the isotopic composition of lead of geological, environmental and biological samples and their potential sources (e.g., Faure and Menssing, 2005; Gulson, 2008; Komárek et al., 2008; Ellam, 2010). This type of plots may additionally help to envisage source apportionment, to visualize contribution of specific sources and to define mixing vectors (e.g., Li et al., 2012). To place insights on these issues in the area, we plotted in the  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{208}\text{Pb}/^{206}\text{Pb}$  diagram of figure 3, the isotopic composition of lead in blood and the available isotopic composition of potential sources. According to this diagram, the isotopic composition of lead in blood would be globally explained in terms of a binary mixing between an end-member represented by the mineralization (and its derivatives) and a more radiogenic end-member represented by bedrock of Morelos and/or Taxco Schist formations, which have distinctive lead isotopic signatures but broadly the same isotopic trend. However, the fact that glazed pottery (made of materials from outside the studied area) and blood show comparable isotopic trends strongly suggests that pottery may be a significant source of lead in the blood and that the variability of the isotopic composition of blood may rather reflect variability of the source of lead in pottery. This statement is consistent with the observations made from the graphs of figure 2, which show that the blood and pottery have nearly identical lead isotopic composition in all isotopic ratios.

As stated before, the lead isotopic composition of indoor dust is highly variable and denotes participation of more than one source. However, around 79% of samples (22 out of 27) have isotopic compositions which are more homogeneous and, within analytical error, identical to those of mineralization and its derivatives. The field distribution of concentrations of dust described before and shown in figure S2, strongly suggests that the tailings are the most significant contributor of lead from dust without underestimating possible minor contributions of the other isotopically identical sources like soils. Some samples of dust have less radiogenic compositions and form a trend toward an unidentified end-member. Rabinowitz (1987) reports that

household interior paints in the United States have a wide range of lead isotopic compositions, which mimics the isotopic composition recorded in our indoor dust samples (Fig. 2). This author and others (e.g., Gulson et al. 2008) demonstrated that interior paints are major contributors to lead of indoor dust and soils around houses and in consequence, a major point source of lead for humans. In this study no house paints were analyzed because recovered paint samples were systematically contaminated by plaster, mud or brick dust. However, the study performed by Kumar (2009) on decorative paints indicates that more than 67% of the Mexican paints contain lead above 90/600 ppm and thus, the participation of interior paints as a source of lead in indoor household dust is very likely. On the other hand since Mexico is the largest importer of decorative paints from United States, it is also probable that interior paints may represent the unidentified, less radiogenic end-member of dust.

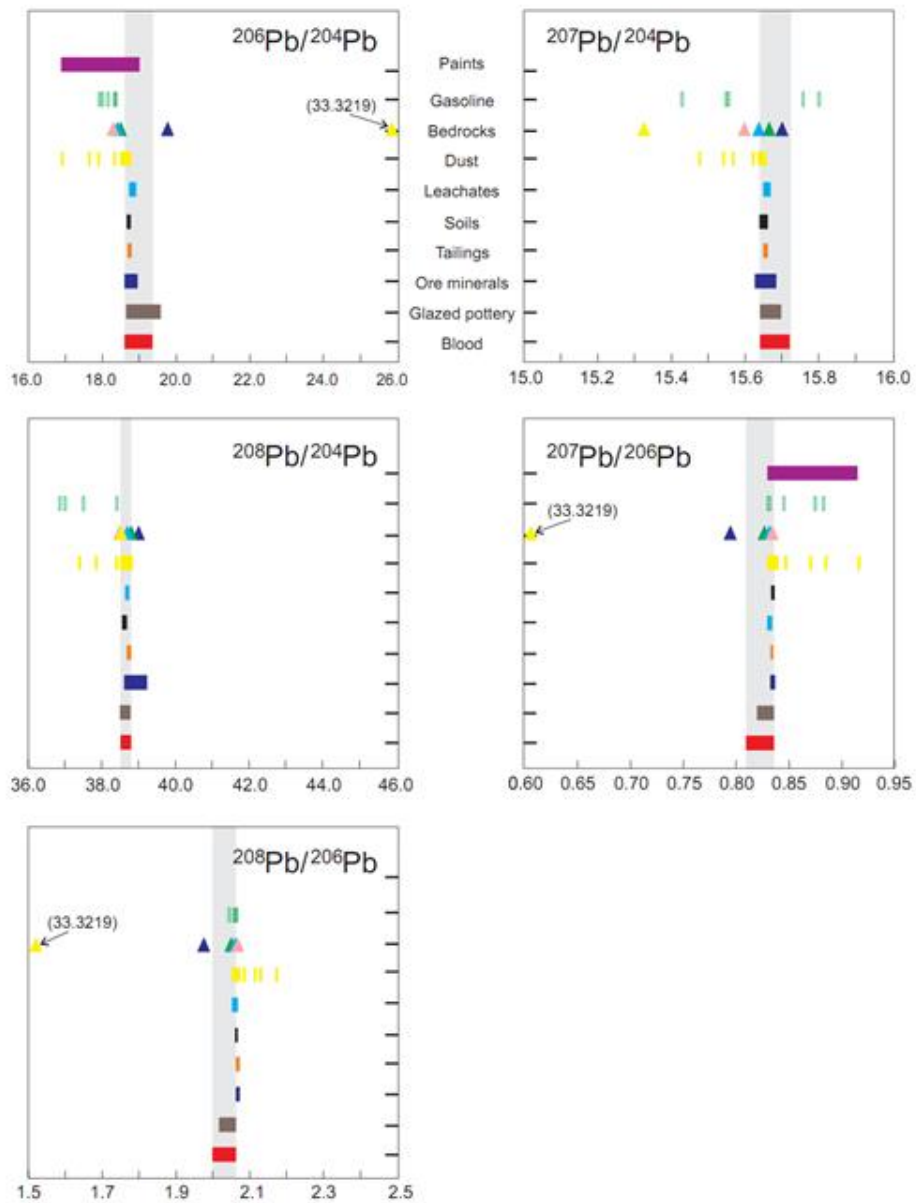


Figure 2. Isotopic ratios of lead in blood of women from the communities of El Fraile and Santa Rosa in the mining region of Taxco, Guerrero, southern Mexico and its comparison with available potential sources. The source of data is indicated in tables S1 to S11 in the data repository.

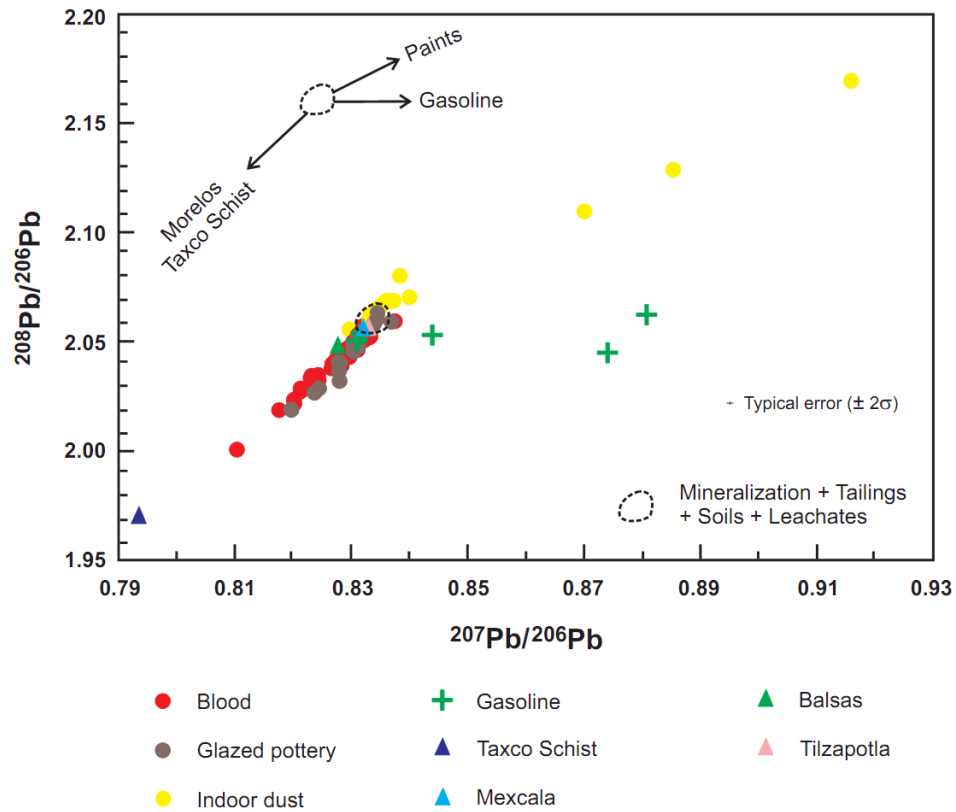
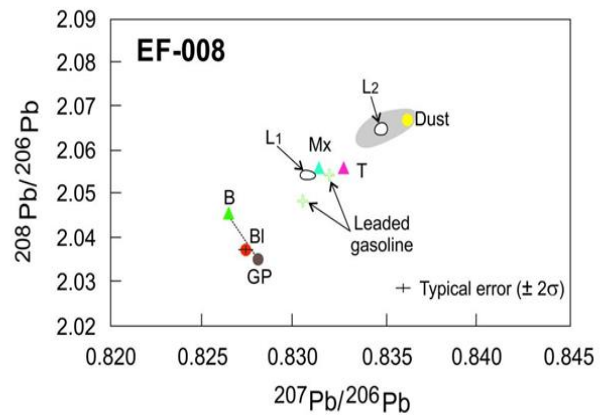
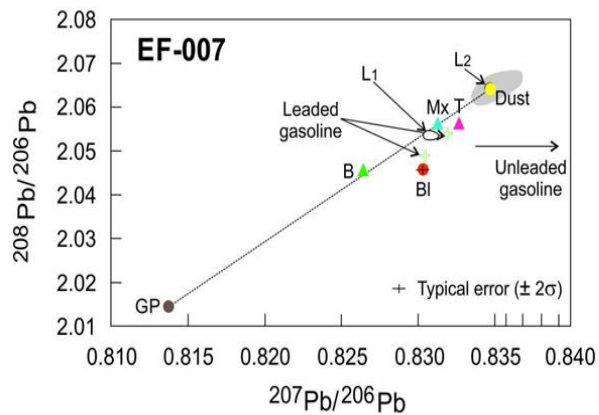
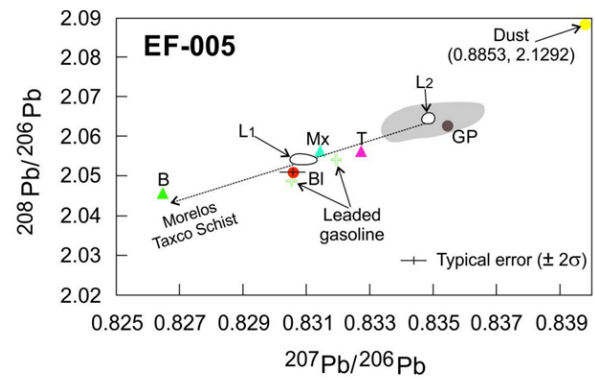
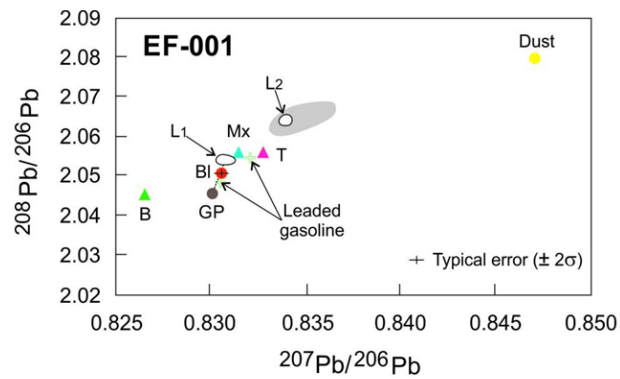


Figure 3.  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{208}\text{Pb}/^{206}\text{Pb}$  plot of women blood and available potential sources in the mining region Taxco, Guerrero (southern Mexico) indicating the field of mineralization and its derivatives (tailings, soils, leachates) and the trend of major sources. The source of data is indicated in tables S1 to S11 in data repository.

In order to identify with a greater degree of certainty the most probable sources, we constructed individual  $^{207}\text{Pb}/^{206}\text{Pb}$  vs  $^{208}\text{Pb}/^{206}\text{Pb}$  plots of the 20 samples of lead in the blood of woman living in houses where indoor dust and glazed pottery samples were also obtained, together with available potential sources (Fig. S4). We stated before that mineralization and its derivatives and glazed pottery appear to be the most significant sources of lead in the blood of the women studied. The analysis of individual graphs indicates, however, that only 7 out of 20 samples representing 35% of plotted samples can be conveniently explained in terms of a simple binary mixing model between these two end-members. Another two samples (EF-007 and EF-037) can also be explained in terms of this mixing model but blood shows a slight displacement to the right of the mixing line suggesting the intervention of a third end-member represented by unleaded gasoline. Eight samples (40%) describe a binary mixing line between the mineralization and a more radiogenic end-member very likely represented by the rocks of the Morelos or Taxco Schist formations with little or no contribution from unleaded gasoline (Fig. S4). Rocks of the Taxco Schist Formation do not outcrop in the studied zone nor underground in mines but rocks of the Morelos Formation host the ore deposits in the zone. In consequence, limestone is a major constituent of the El Fraile and La Concha tailings deposits and thus, Morelos rocks represent the more plausible radiogenic, end-member. Only sample EF-008 has an isotopic composition nearly matching that of its corresponding glazed pottery; sample EF-001 shows an intermediate isotopic composition between that of glazed pottery and mining leachates; and, sample SR-010 shows influence of mineralization and unleaded gasoline (Fig. S4).

Arcega-Cabrera et al. (2010) demonstrated that tetraethyl lead derived from leaded gasoline before 2000 is still ubiquitous in the zone. The isotopic composition of Mexican leaded gasoline reported by Martinez et al. (2004) and Morton-Bermea (2011) is very similar to that of some lithologies in the zone (Tables S9 and S10; Fig. S4) and in consequence, contribution of residual leaded gasoline to lead in blood from the studied women cannot be neglected.



Grey field = mineralization + tailings + soils; L1 = family I leachates; L2 = family II leachates; BI = Blood; GP = Glazed pottery; Mx = Mexcala Fm; B = Balsas Fm; T = Tilzapotla Fm.

Figure S4. Individual  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{208}\text{Pb}/^{206}\text{Pb}$  plots of lead in the blood of women of reproductive age from El Fraile and Santa Rosa in the mining region of Taxco, Guerrero southern Mexico showing their relationships with available potential sources in the zone. Dashed line indicates the proposed mixing lines to explain the origin of lead in blood. The sources of data are indicated in tables S1 to S11.

Figure S4. (Continuation)

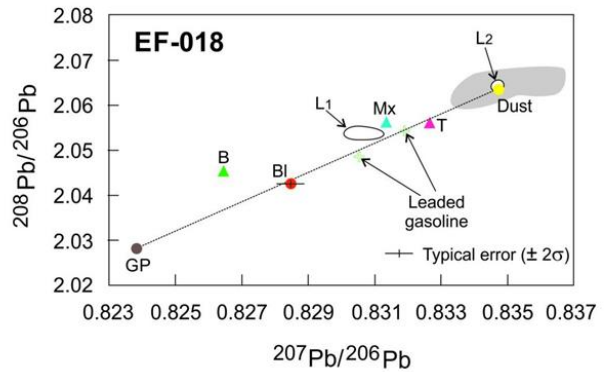
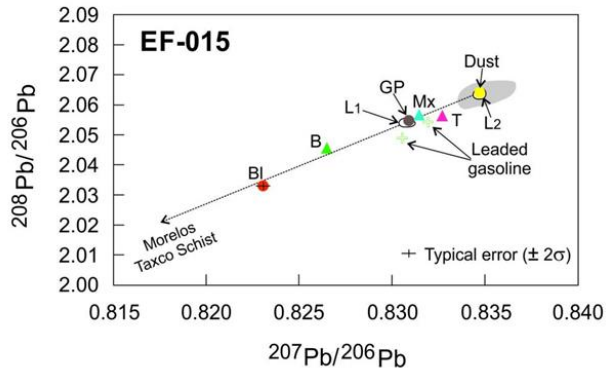
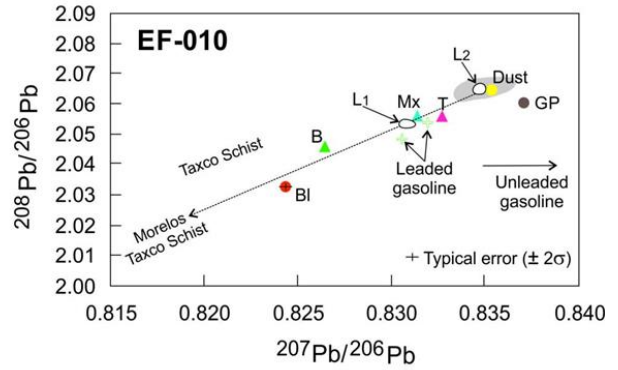
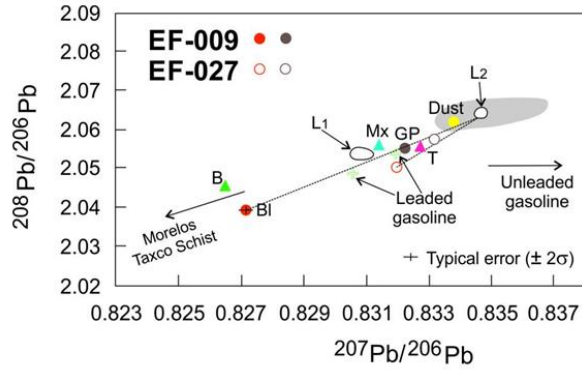




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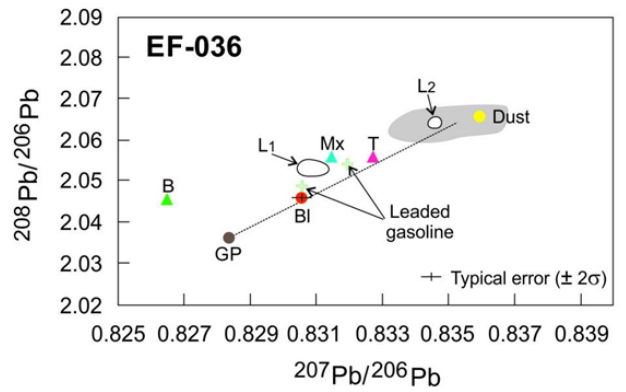
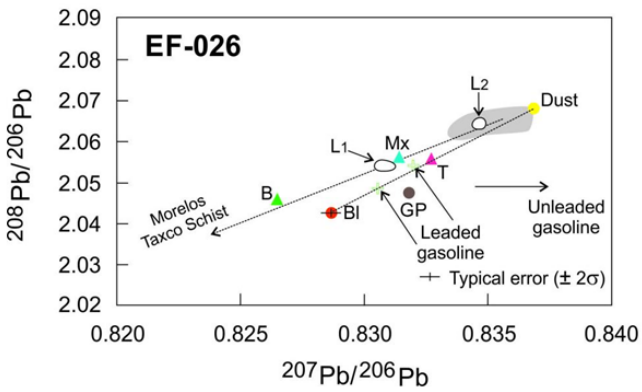
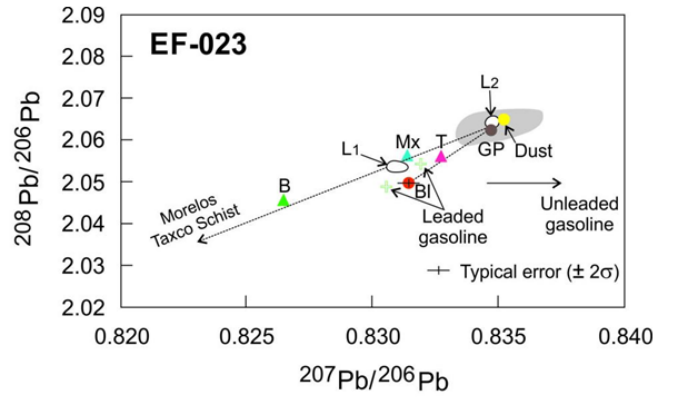
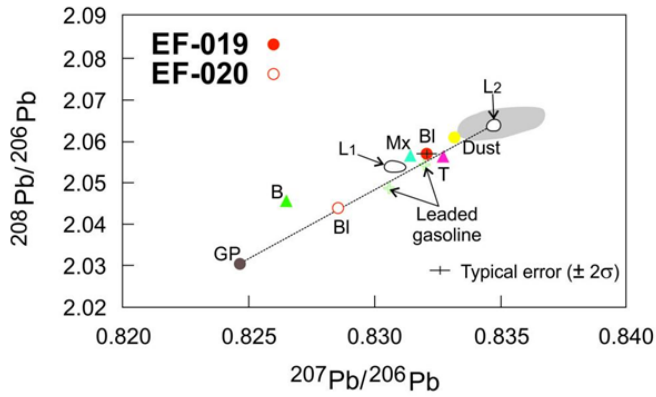


Figure S4. (Continuation)

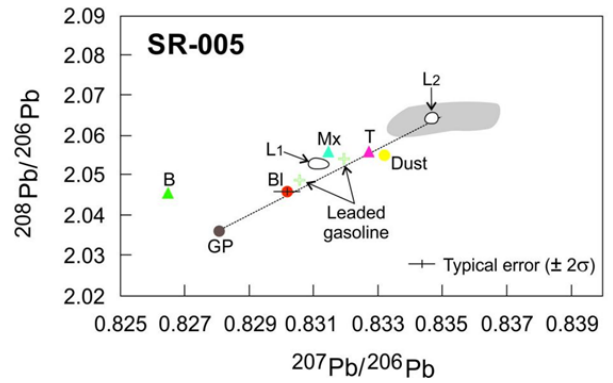
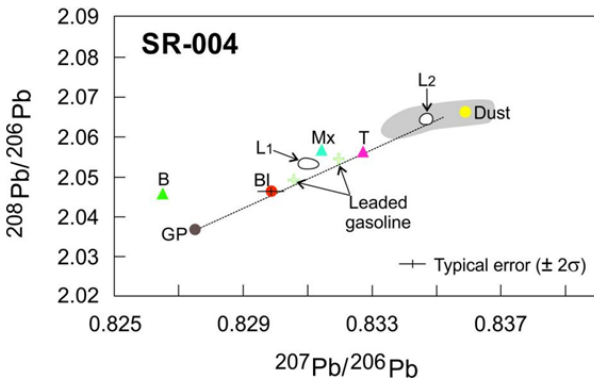
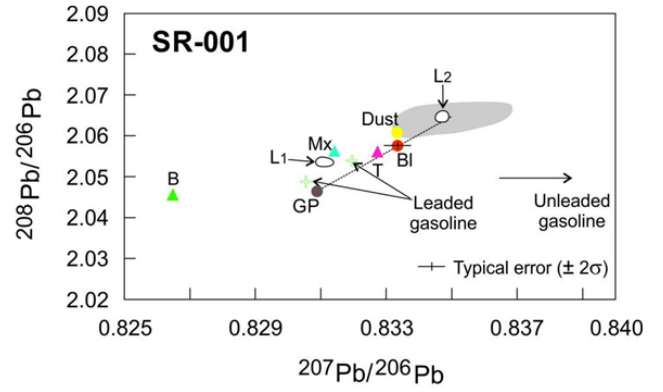
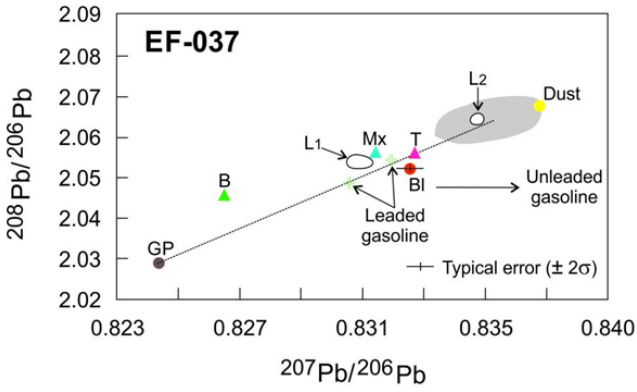
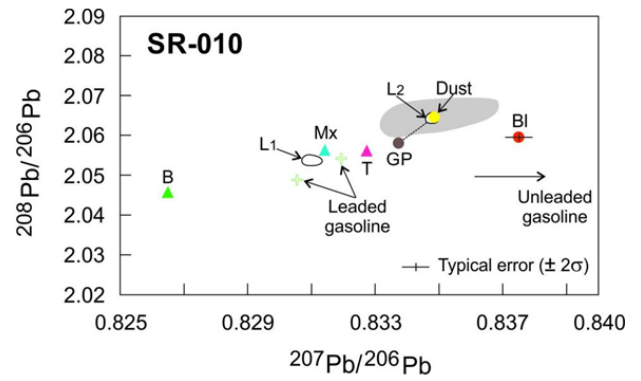
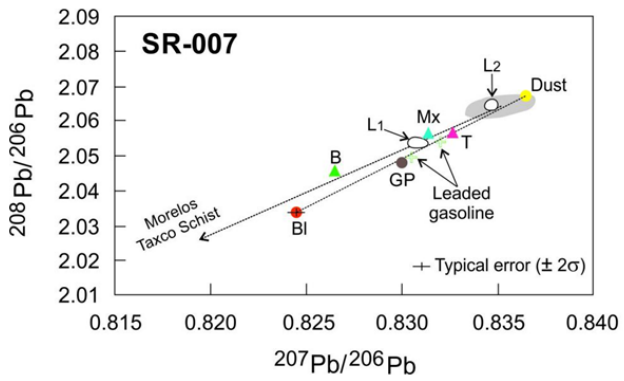


Figure S4. (Continuation)



## 6. Conclusions

Women of reproductive age (15 to 45 y) from the rural communities of El Fraile and Santa Rosa of Taxco, Guerrero in southern Mexico are exposed to multiple sources of lead from both natural and anthropogenic origin. Natural emitters are restricted to bedrock whereas anthropogenic sources are varied and include glazed pottery, tailings, mine-impacted soils and dust, mining leachates, interior paints, and leaded and unleaded gasoline. Lead concentrations of blood indicate that about 36 % of the studied women have concentrations above  $10 \mu\text{g dL}^{-1}$  and up to 87 % above  $5 \mu\text{g dL}^{-1}$ . Levels of lead in indoor dust are largely variable but only 15 % of the samples are above the Mexican Regulatory Guideline for urban soils ( $400 \text{ mg kg}^{-1}$ ). By contrast, 85 % of glazed containers used for cooking or food storage are well above the maximum  $2 \text{ mg L}^{-1}$  of soluble lead established in the Mexican Guideline for leachable lead in glazed pottery.

Soils and most indoor dust have isotopic compositions that mimic that of mineralization, tailings and leachates confirming widespread dispersal of lead and associated toxic metals from ores and mine wastes to neighboring natural resources. The lead isotopic data indicate that mineralization (and its derivatives), glazed pottery and the Morelos bedrock are the most significant sources of lead in the blood of women in the zone. Unleaded gasoline seems to have a minor contribution in many women but the influence of residual leaded gasoline could not be demonstrated. Paints seem to be a major contributor to lead in some samples of indoor dust but their influence is not recorded in any blood sample.

Even though the application of isotopic ratios of lead for identifying point sources and their apportionment in diverse environmental matrices of geogenic origin (i.e. water, soils, sediments, dust) has been successful, their application in biological materials and particularly in human fluids and tissues is still limited (e.g., Gulson, 2008). Due to their quotidian activities and dietary habits, humans are exposed to a larger number of point and diffuse lead sources of both, natural and anthropogenic origin relative to geogenic samples. This fact increases variability in the isotopic composition of human fluids and tissues, which in turn results in an increase in the

number of individuals and environmental materials under investigation to better register variability and to assign contribution from specific sources. Our study supports the application of lead isotopic ratios in identifying potential sources and their apportionment in the blood of women exposed to multiple sources of both natural and anthropogenic origin.

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# Appendix



**ANEXO I**  
**UNIVERSIDAD AUTÓNOMA DE GUERRERO**  
UNIDAD ACADÉMICA DE CIENCIAS QUÍMICO BIOLÓGICAS  
UNIDAD ACADÉMICA DE CIENCIAS DE LA TIERRA

**CARTA DE CONSENTIMIENTO INFORMADO**

FOLIO \_\_\_\_\_

**Proyecto: Mujeres y Metales en Taxco de Alarcón, Gro.**

Al firmar este documento, doy mi consentimiento para que me entreviste un miembro del personal del **Laboratorio de Toxicología y Salud Ambiental** de la Unidad Académica de Ciencias Químico Biológicas, ubicada en Av. Lázaro Cárdenas s/n, Chilpancingo, Gro., o **del Laboratorio de Geoquímica** de la Unidad Académica de Ciencias de la Tierra, ubicada en Ex hacienda de San Juan Bautista s/n en Taxco el Viejo, Gro., de la Universidad Autónoma de Guerrero. Entiendo que seré entrevistada, en los horarios que se me indiquen o que así convengan, se me harán preguntas sobre los antecedentes personales. También se me realizará una toma de muestra sanguínea por punción venosa suficiente para los análisis de la investigación, una muestra de orina y proporcionaré una muestra de barro y polvo. La entrevista durará aproximadamente 20 minutos, también estoy enterada que el entrevistador(a) puede ponerse en contacto conmigo en el futuro, a fin de obtener más información. Se me ha dicho que las respuestas a mis preguntas y demás información obtenida no serán reveladas a nadie y que en ningún informe de este estudio se me identificará jamás en forma alguna. Sé que tengo la decisión de abandonar el estudio cuando lo desee y que además con las intervenciones a las que seré sometida no corro ningún peligro en salud. Como compensación por cualquier esfuerzo o molestia relacionados con mi participación en este estudio me serán proporcionados los resultados de los análisis practicados, sin costo alguno.

Para cualquier duda podré comunicarme al Laboratorio de Toxicología y Salud Ambiental de la Unidad Académica de Ciencias Químico Biológicas de la UAG. Teléfono 4725503 con la Dra. Ma. Elena Moreno Godínez o al Laboratorio de Geoquímica de la Unidad Académica de Ciencias de la Tierra de la UAG. Teléfono 7626213466 con el Dr. Oscar Talavera Mendoza.

\_\_\_\_\_/\_\_\_\_\_/\_\_\_\_\_

Fecha

\_\_\_\_\_

Nombre y firma de entrevistado(a)

\_\_\_\_\_

Nombre y firma del entrevistador (a)



**ANEXO II**  
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FOLIO \_\_\_\_\_

**CUESTIONARIO**

**Proyecto: Mujeres y Metales en Taxco de Alarcón, Gro.**

**Nombre del encuestador:** \_\_\_\_\_

**Fecha de la entrevista:** día \_\_\_\_\_ mes \_\_\_\_\_ año \_\_\_\_\_

**NOTA IMPORTANTE: Solo Podrá Ser Contestado por el Paciente**

**1.-FICHA DE IDENTIFICACIÓN DEL PACIENTE**

1.1. Nombre \_\_\_\_\_

Talla: \_\_\_\_\_ Peso: \_\_\_\_\_ Presión arterial: \_\_\_\_\_ IMC: \_\_\_\_\_

1.2. Edad (años) \_\_\_\_\_ meses \_\_\_\_\_

1.3. Inicio de menarca (fecha): \_\_\_\_\_

1.4. Número de embarazos: \_\_\_\_\_ Número de abortos: \_\_\_\_\_ Número de hijos: \_\_\_\_\_

1.5. A qué edad tuvo su primer hijo: \_\_\_\_\_

1.6. Domicilio actual: \_\_\_\_\_

1.7. Localidad: \_\_\_\_\_ Municipio \_\_\_\_\_

1.8. Lugar de nacimiento \_\_\_\_\_ Municipio \_\_\_\_\_

**2. CARACTERÍSTICAS SOCIOECONÓMICAS DE LA PACIENTE**

2.1. ¿Hasta qué año estudió? \_\_\_\_\_ Profesión, si aplica: \_\_\_\_\_

2.2. ¿Sabe leer? \_\_\_\_\_ 2.3. ¿Sabe escribir? \_\_\_\_\_ 2.4. ¿Leer y escribir? \_\_\_\_\_

2.5. ¿Cuál es el ingreso promedio mensual de la familia? \$ \_\_\_\_\_

2.6. La casa en que vive es: Propia \_\_\_\_\_ Rentada \_\_\_\_\_ Prestada \_\_\_\_\_

2.7. ¿Cuántos cuartos tiene su casa? \_\_\_\_\_

2.8. ¿Cuántos dormitorios tiene su casa? \_\_\_\_\_

2.9. ¿Cuántas personas viven en su casa? \_\_\_\_\_

2.10. ¿De qué material está construida su casa?

Ladrillo y cemento \_\_\_\_\_ Adobe \_\_\_\_\_ Madera \_\_\_\_\_ Lámina o cartón \_\_\_\_\_



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2.11. El piso de la casa es de:

Tierra apisonada\_\_\_\_\_ Cemento u otro\_\_\_\_\_

2.12. ¿Con qué servicios cuenta su domicilio?

Luz\_\_\_\_\_ Drenaje\_\_\_\_\_ Agua potable\_\_\_\_\_ Tel\_\_\_\_\_

2.13. ¿Cómo eliminan la excreta?

Sanitario WC\_\_\_\_\_ Letrina\_\_\_\_\_ Fosa séptica\_\_\_\_\_ Suelo\_\_\_\_\_

**3. EXPOSICIÓN**

**3.1. HISTORIA DE RESIDENCIA**

Siempre ha vivido en la comunidad (Marque una **X** en la línea y omita el cuadro siguiente) \_\_\_\_\_

Fecha (en años)	Localidad	Municipio	Estado
De a			
De a			
De a			
De a			

3.2. ¿Qué tipo de agua utiliza para beber y/o cocinar? **MARQUE CON UNA X** y anote el tiempo de uso.

USO DEL AGUA	ENTUBADA DE JALES	POZO	NORIA	HIDRANTE PÚBLICO	GARRAFÓN
Beber					
Cocinar					
Actividad del hogar y aseo personal					

3.3. ¿En los últimos 7 días ha comido pescado, mariscos o champiñones (Incluyendo los de lata)?

Sí \_\_\_\_\_ No\_\_\_\_\_

3.4 ¿En qué lugar permanece más tiempo durante sus actividades cotidianas?

Casa: \_\_\_\_\_ Escuela: \_\_\_\_\_ Trabajo: \_\_\_\_\_ Otro especifique: \_\_\_\_\_



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**4. CONFUSORES**

4.1. ¿Sale o esta mucho tiempo expuesta al sol?

Sí\_\_\_\_\_ No\_\_\_\_\_ ¿Cuántas horas?\_\_\_\_\_

4.2. ¿Ha tomado antibiótico en las últimas dos semanas?

Sí \_\_\_\_\_ No\_\_\_\_\_ ¿Por cuantos días?\_\_\_\_\_

4.3. ¿Ha tomado o toma calcio? Sí\_\_\_\_\_ No\_\_\_\_\_ ¿Por cuantos días?\_\_\_\_\_

4.4. ¿Ha tenido asma, alergias, rinitis, conjuntivitis frecuentes? Sí\_\_\_\_\_ No\_\_\_\_\_

4.5. ¿Tiene o ha tenido infecciones parasitarias diagnosticadas como amibas o lombrices recientemente (2 semanas antes)? Sí\_\_\_\_\_ No\_\_\_\_\_

4.6. ¿Utiliza Ud. Loza de barro que tiene capa brillante por dentro? Sí\_\_\_\_\_ No\_\_\_\_\_

4.7. Fuma: Si \_\_\_\_\_ No: \_\_\_\_\_

4.8. ¿Cuántos cigarrillos por día?\_\_\_\_\_ ¿Qué marca de cigarrillos?\_\_\_\_\_

4.9. ¿Toma bebidas alcohólicas? Sí: \_\_\_\_\_ No: \_\_\_\_\_

4.10. ¿Con que frecuencia?\_\_\_\_\_

4.11. ¿Convive con algún fumador, o algún miembro de la casa fuma? Sí\_\_\_\_\_ No\_\_\_\_\_

4.12. ¿Cuántos cigarrillos por día?\_\_\_\_\_ ¿Qué marca de cigarrillos?\_\_\_\_\_

4.13. ¿En dónde trabaja su esposo y/o su padre?

Minería\_\_\_\_\_ Campo\_\_\_\_\_ Otro (especifica) \_\_\_\_\_

4.14. Si trabaja en el campo ¿Qué actividad realiza?\_\_\_\_\_

4.15. ¿Usa plaguicidas? Sí\_\_\_\_\_ No\_\_\_\_\_ ¿Cuáles?\_\_\_\_\_

4.16. ¿En dónde trabaja usted? Hogar\_\_\_\_\_ Minería\_\_\_\_\_ Campo\_\_\_\_\_ Empleada

(especificar) \_\_\_\_\_ Otro especificar) \_\_\_\_\_

4.17. ¿En su casa tiene algún taller para el trabajo de la plata, alpaca u otro metal?  
Especifique\_\_\_\_\_

4.18. Alguien de la casa realiza algún trabajo de artesanía:

Especifique\_\_\_\_\_

Además de sus actividades desempeña algún trabajo como:

4.19. ¿Ayudar en la siembra aplicando fertilizantes y/o insecticidas? Sí \_\_\_\_\_ No\_\_\_\_\_



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**4.20.** ¿Ayudar en algún taller de platería o artesanal? Sí \_\_\_\_\_ No \_\_\_\_\_

Otro \_\_\_\_\_ Especificar \_\_\_\_\_

**4.21.** Usted tiene o ha tenido regularmente alguno de estos síntomas:

01 Temblor de brazos o manos \_\_\_\_\_

02 Temblor de piernas \_\_\_\_\_

03 Calambre en piernas \_\_\_\_\_

04 Pérdida de memoria momentánea o persistente \_\_\_\_\_

05 Prurito constante en todo su cuerpo \_\_\_\_\_

06 Dolores musculares frecuentes \_\_\_\_\_

07 Dolor de huesos \_\_\_\_\_

08 Problemas renales \_\_\_\_\_

09 Dolor de cabeza frecuente \_\_\_\_\_

10 Infecciones Urinarias \_\_\_\_\_

11 Callosidades en manos, pies, o cuerpo en general \_\_\_\_\_

12 Diabetes \_\_\_\_\_ tiempo de diagnóstico (años) \_\_\_\_\_

13 Hipertensión \_\_\_\_\_

**4.22. ANTECEDENTES PERSONALES PATOLÓGICOS INCLUYENDO SINTOMATOLOGÍA FRECUENTE:**

\_\_\_\_\_  
\_\_\_\_\_

**4.23. ANTECEDENTES FAMILIARES:**

Mamá: \_\_\_\_\_

Papá: \_\_\_\_\_

Abuelos: \_\_\_\_\_

Otros (hijos, hermanos, etc): \_\_\_\_\_







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FOLIO \_\_\_\_\_

**CUESTIONARIO II**

**Proyecto: Mujeres y Metales en Taxco de Alarcón, Gro.**

**Nombre del encuestador:** \_\_\_\_\_

**Fecha de la entrevista:** día \_\_\_\_\_ mes \_\_\_\_\_ año \_\_\_\_\_

1.- ¿De dónde proviene la loza de barro que utiliza?

- a) Lugar en el que radica    b) Vendedores ambulantes    c) Otro lugar Especifique \_\_\_\_\_

2.- ¿Qué utensilios de barro utiliza?

- a) Jarros    b) Cazuelas    c) Platos    d) Todo lo anterior

3.- ¿Cuánto tiempo tiene el utensilio?

- a) 1-6 Meses    b) 6 meses a un año    c) De 1 año-5 años    d) Más de 5 años

4.- ¿Guarda alimentos en utensilios de barro?

- a) Sí    b) No

5.- En el utensilio de barro se guardan alimentos:

- a) Sólidos    b) Líquidos    c) Ambos

6.- ¿Usa los utensilios de barro como vajilla?

- a) Sí    b) No

7.- ¿Con que frecuencia?

- a) Diario    b) 1-3 veces por semana    c) 1 vez al mes    d) Solo en eventos

8.- ¿Cocina en utensilios de barro?

- a) Sí    b) No

9.- ¿Con que frecuencia?

- a) Diario    b) 1-3 veces por semana    c) 1 vez al mes    d) Solo en eventos

10.- ¿Qué alimentos se cocinan con mayor frecuencia?

- a) Frijol    b) Arroz    c) Sopa    d) Jugos    e) Café    f) Mole    g) Otros especifique: \_\_\_\_\_