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Mercurialism Determination in Fetuses Bone Remains from Toluquilla, Queretaro, Mexico

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

There are few mentions in Mexican archaeology for study of prehispanic underground mining to exploit the cinnabar, but there are lots of references about their use as pigment for different kind of artifacts in Mesoamerican archaeology. Our research settled at Sierra Gorda emphasized the ancient life form specialized in production and trade of mercury ore, and that includes specific analysis to the human remains gathered in the archaeological work to determination how many of their inhabitants were evolved at this economic activity.

The ancient mining was so intensive and extensive at this region pollutes their environment affecting to the ancient population in their health. We present a few rare and special cases with important medical implications about health condition of ancient pregnant women with a journey through toxicological issues and bioaccumulation of mercury in their bodies.

The archaeological excavation provides skeletal remains of 200 individuals. We presented 37 cases that represent the diversity of population composition with adults (men and woman), adolescents and infants and 4 rare cases of fetuses buried at Toluquilla archaeological zone. In

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each case a bone sample was taken for determination of heavy metals by spectrometric chemical techniques.
The measurement results for total mercury in bones lead us evaluate the medical implications about their individual health condition and propose a few lines concerning the ancient public health.

Keywords: Mercurialism; cinnabar; toxicity; bioaccumulation; health pregnancy; mining; prehispanic bones; Sierra Gorda; Queretaro.

1. INTRODUCTION

Today mining activity, mostly open-air exploitation, that management has left clear of a significant amount of heavy metals involves the processes of extraction of minerals. With the purpose of wide production, either because they used in the process of benefit, as it is the case of the cyanidation, a concern and awareness about effects of metals on the environment and the human population has increased.

The issue is not new, for years, it is used for various purposes-heavy metals and safety measures were less careful. At the south of the Sierra Gorda in north portion of Queretaro state - about 400 kilometers from the city of Mexico (see Fig. 1), - it has been known the use of Mercury Red sulfide powder, known as cinnabar. The Cinnabar was used in ancient times as a symbol of wealth and power. The pigment was the Earth's blood in the ancient belief system, or pigment of life, the elemental colour of the underworld and drug because their inhalation or use in rituals cause altered states of mind. The quicksilver was the magic liquid, is the natural liquid that don't wet and the perfect mirror.

If we consider that authors such as Santiago Español [1] emphasize that a toxic or contaminant is every alien element to the human body or its amount greater than the natural background, which was established since ancient times and its effects were known later 1473 as occupational hazard [2].

With that, it became clear that exposure to heavy metals such as mercury is a phenomenon of public and occupational health, when exceeds the maximum levels that occur in nature and comes to produce a disease (see [2-7]). In addition, the difference in the availability of metals in nature should be considered when ore deposits exist and the native or "background" value is higher than that in a region without ore deposit. In the Sierra Gorda of Querétaro State there are ore deposits of various metals formed by hydrothermal phenomena of its geological formation, and cinnabar or mercury had been used from 200 BC to the present day. (see [8,9])

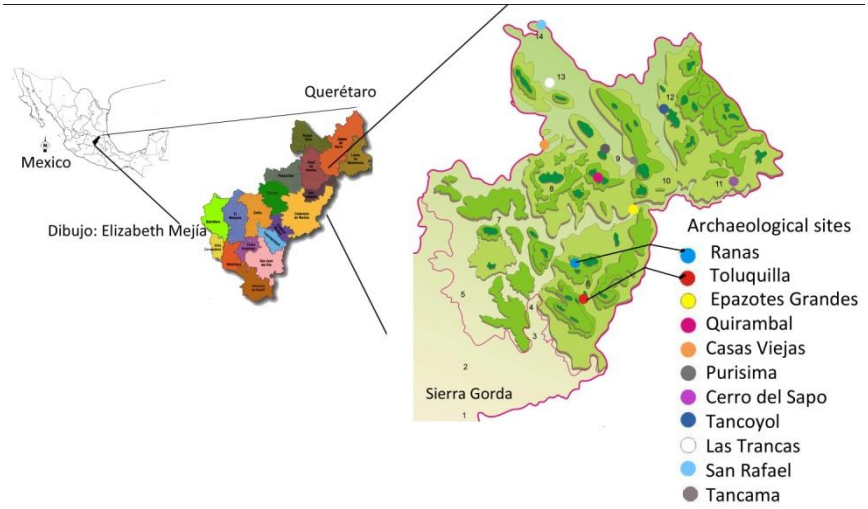

Fig. 1. Map of Mexico (left), Queretaro state (center) and Sierra Gorda (right)

2. MERCURY FORMS, TOXICOLOGICAL DYNAMICS

Mercury is highly toxic, and this toxicity depends on the ways of penetration into the organism, individual metabolic rate, excretion rates and the synergistic or antagonistic effects of other agents [1], [3], and the concentration in the different bodies under a high flows of blood storage, biotransformation [2], [1] and excretion [1,3,5].

There are organic and inorganic mercury, and each one has special kinetic toxic characteristics and toxicological properties inherent in their status. For example, the toxicity of organic mercury and methyl mercury is higher than that of elemental mercury and inorganic compounds [6].

There are factors to be considered in the study of metals in ancient or modern population. The various forms of mercury and their toxicological properties vary in each case, the elemental mercury [e-Hg], is lipid soluble, diffusible in membranes and it is oxidized intracellularly. While inorganic mercury [i-Hg], is water soluble and less diffusible through membranes. This can induce protein synthesis type of the metalotionina in the kidney, being the main union of mercury to proteins; the alkyl-mercury compounds (al - Hg), mainly the methyl mercury (me-Hg), are lipid-soluble, highly diffusible through membranes and are slowly transformed into i-Hg. In addition, organic mercury compounds (or-Hg) and (alox-Hg) are soluble in lipids and quickly degradable in the body to i-Hg [2,1]. All of them in a high degree of concentration are deposited in bones, being the center of the long bones and teeth, both are the type of samples with less susceptibility to the surrounding soil contaminants.

2.1 Pollution

The entrance of mercury to the body occurs through respiratory or digestive tracts and skin. The airway is by: a) mercury vapor in the elementary form, present in gas traps, easily penetrates the membrane of the pulmonary alveoli and passes into the blood, which absorbed 80% of inhaled quantity. (b) when breathing contaminated dust. Both paths are the most important routes of entry in the mining activity [3,2].

The path through digestive tract of elemental or metallic mercury occurs from swallowing water, animals and/or food containing metals. This is the ingestion of contaminated water, fish or other marine animals; and plants or other animals contaminated with toxic dust [3,2]. Its absorption is in quantities of less than 0.01%, although the inorganic mercury has absorption of approximately 7% (Español, S. 2001: 10) [1]. This is the most important in the case of methyl mercury and therefore is the most important input in the field of public health.

The dermal route is not well known since it is unknown exactly the amount of mercury or its compounds that can be absorbed through the skin, and it is not feasible that this absorption pathway have an important role in comparison with those previously identified. If anything, the presence of mercury in the hands could be relevant insofar as these salts could be put in contact with mucous membranes (mouth, nose, etc.) that absorb mercury more easily [1]. It is estimated that the normal content of mercury in the human body varies between 1-13 mg/kg, of which the methyl mercury is equivalent to 10% of the total content.

2.2 Elimination

It is known that the body efficiently removes mercury in about two weeks, though, when intake exceeds the elimination or even when there are difficulties in excretion concentration reaches the critical point causing toxic effects. Toxic substances can be disposed of without any changes in the body, however it is not uncommon that a substance be biochemically altered to make them water-soluble or fat-soluble and thus be more easily filtered by the kidney tubules and excreted in the urine. Failure to do so would mean that the excretion occurs in the feces, and to a lesser extent by sweat and saliva. Interestingly, in the transformation process that precedes the excretion some substances can become toxins or reduce their toxicity [2].

In any case, biotransformation aims to eliminate toxic substances from the body or otherwise converted into neutral or less harmful substances that can be stored in fatty tissue. This process comprises two phases; the first is oxidation, reduction and hydrolysis of the toxic substance and the second stage combines with bile salts. The most important bodies in the transformation of mercury are the kidneys, lungs, bowel and brain [2].

The contribution of each path to the total elimination of mercury depends on the type of mercurial compound and occurs at different times after exposure. The fecal route is the most important in the elimination of methyl mercury since 90% of this salt is unchanged after exposure. Other routes of excretion that must be taken into account are: saliva, hair, sweat and exhaled air.

In the case of substances whose toxicity coincides with a stable state of ingestion / elimination, the clinical manifestations of toxicity vary over time that elapses after the exhibition-giving rise to acute, sub-acute and chronic intoxication. The latter is the most common and constitutes the so-called "Hydrargyrisme or Mercurialism", which manifests itself differently in the poison elemental or inorganic mercury than those in which the causative agent is methyl mercury.

In terms of its effects, the mercury occurs in two clearly defined stages, first period of impregnation or absorption, which is characterized by symptoms little precise and non-specific, ranging from lack of appetite, tiredness, weight loss, headaches, difficulties to reconcile sleep, pain in arms and legs. The second phase is the period of intoxication where there are disturbances in digestive tract, eye, kidney, skin and nervous system [1].

In the case of organic compounds of mercury (methylmercury) poisoning, the clinical picture differs from the reflected above. This poisoning does not present the classic symptoms, except the tremor, and even if the level of mercury in urine is insignificant. They are heart disorders, paralysis, and death may ensue due to infectious complications. We must start from the premise, that a toxic as mercury produces irreversible lesions at the level of the nervous system.

In modern medicine the treatment of poisoning by mercury includes the use of antidotes that reduce the amount of mercury in the host tissues, either forming an inactive complex with mercury or facilitating the elimination of the metal out of the tissues. Antidotes, naturally, is complemented by general supportive therapy. [1]

In chronic exposure for a long period of divalent mercury, it is first deposited in mitochondrial cells in the kidneys and liver; while the distribution of methylmercury is more uniform. After an ongoing mercury takings most goes to the brain, liver and kidney; but it has been detected also in epithelium of the thyroid, the cells of the spinal cord and the adrenal glands, in the spermatocytes, in pancreatic epithelium, the epidermis, in the Crystal, hair and blood.

According to specific works of current populations, there is a constant and proportional relationship of pollution between hair and blood in chronic exposures by the consumption of contaminated food, specifically of fish, the correlation of Sherlock et al. is mercury in hair = 0.367 x Hg blood + 0.694[1]. While authors such as Hansen J. et al. reported that the correlation between concentration in hair and blood is 0.9222[2]. (see [10,11])

3. POLLUTION OF MERCURY IN PREGNANT WOMEN

The objective of our research is known how pre-Hispanic mining affects to the ancient population, particularly to children. Analyzing the result of metals according to age and sex in the pre-Hispanic population of fetuses are particularly important since they reflect their health and that of the mother, since after the poisoning, maternal contaminants pass through the placental barrier and it accumulates in the fetal liver after their oxidation (Repeto, 1995: 377). Authors scoring: when there is mercury in the body the relationship between hair and blood is r mercury = 0.6589, while methylmercury is r = 0.7348. (See [7], [6], [4], [5] and [2]).

In poisoning by methyl mercury after crossing the placental barrier [6], it affects the fetus since it depresses the cholinergic system due to its inhibitory action of acetylcholine transferase, both as the striate core in the cerebral cortex. Analysis has been in animal experiments in the decrease in motor activity, rotational dysfunction and hyperthermia. Methylmercury inhibit the entrance of dopamine, serotonin and norepinephrine (in that order) in the striated synaptosomes, hypothalamic and cortical respectively. High doses of methylmercury (15 - 50 mg/kg) decreased the synthesis of dopamine, interference by cholinergic mechanisms of neurotransmitters (Repeto, 1995: 381) [7]. This alters the formation of platelets, release and platelet aggregation. In erythrocytes it decreases

[1] *Hg in hair = mg/Kg and Hg in blood = µg/kg, with a correlation coefficient is 0.837.*

[2] *Represented by the following equation: Mercury in hair = 287 x Hg blood + 63.4 and Hg in hair = µg/Kg, Hg in blood = µg/kg is equal to r = 0.9222.*

the concentration of zinc and magnesium and neutrophils, increases iron concentration and decreases stress tolerance and sexual activity by inhibition of testicular and adrenal steroids (Repeto, 1995: 382). [7]

When pregnancy goes this way and the birth comes to term, residual effects of uterine exposure to mercury are the same as that are manifested in the cerebellum of mice exposed after a period of 10-12 week after apparent birth in leaves cerebellar and simplified lateral ventricles, caudate nucleus and putamen reduced.

Uterine exposure to residual pathologic effects are manifested by focal areas of astrogilosis or astrocytosis in the molecular layer, in residual bodies, dendrites of granule on neurons and there are degenerative changes in myelinated axons. By this the consequences are varied since the product may present hydrocephalus, the decrease of the cerebral cortex in parietal section, increasing the thickness of the hippocampus in the occipital section and modifies the production of dopamine and noropinefrina, which means the alteration of the synaptic dynamic evolved in neural development of centrals catecholaminergic [7].

Repeto [7] concerns the case of Minamata, Japan, where the greatest concentration of methylmercury intoxication-related is 5 mg/kg, value that corresponds to 800 µg/L in blood and 200 mg/g in hair. The concentration of 1 - 2 mg/kg in brain (200 ng/g blood) is the lowest concentration at which neurological signs are observed [12]. In studies of Bakir et al. [13] for population contaminated in Iraq, observed the relationship between the body load and rate response frequency.

Thus, for the sign paresthesia, the minimum value of the body burden is 25-40 mg of mercury, which corresponds to a concentration of 250-400 µg/L. Nordberg and Strangert [14] suggested that the first effect in 5% of the adult population can be expected after long periods of daily intake of methylmercury between 3-7 µg of mercury/Kg of body weight, resulting in a concentration of 200 µg/L in blood and a maximum between 50-125 µg of Hg/g of hair. According to this, an intake of 50 µg/day in adults involves a risk of 0.3% of the paresthesia symptom and for an intake of 200 µg/day the risk is 8% (WHO, 1990) [15].

In prenatal exposure doses neurological effects are dependent on a range from elusive changes in brain function in low doses, until a severe neurological syndrome with changes in brain structure in high exposure. In mothers who have a concentration in 5-10 µg/g hair, they have mild retardation and when ranges from 10 to 20 µg/g have a 5% risk, and above 70 µg/g implies a serious risk (30%) of neurological disorders of the fetus (Repeto, 1995: 386). [7]

4. PRE-HISPANIC POPULATION OF TOLUQUILLA

The study population was prehistoric skeletons excavated for 12 years (1993-2005) of 25 burials from Toluquilla archaeological site (see Picture 1), located at south of the Sierra Gorda, in the State of Querétaro, about 400 kilometers Northwest of the city of Mexico [16]. In the Toluquilla's archaeological project despite having about 200 skeletons were only 32 including adult men and women (12), adolescents and infants (16) and 4 fetuses to determine 11 metals (Hg, As, Li, Cd, Cr, Cu, Ni, Pb, Se, Zn, Sb) by PIXE and Atomic absorption technique performed by the laboratory of Geo sciences of the National Autonomous University of Mexico Campus Juriquilla in Queretaro, in charge of Dr. Gilberto Hernández. Working together was held from 2002 to 2009 [8], today Juriquilla's team analyses the soil, geological and environmental aspect and we have devoted to the analysis of the people bone's related to results mines activities, buildings and archaeological sites. [9] (see Fig. 2).

There are no soft organs in our population and we had to evaluate and compare the contents of native soil metals or 'background value' to find out if they are not transmitted and if indeed they bioaccumulate in living individuals. So fetuses were 4 cases and 16 infants, wich were analyzed for heavy metals in prehistoric bones, they are of particular interest since it is also an indirect indicator of the health of the mother who polluted their fetuses through a placenta.

5. RESULTS AND DISCUSSION

The following skeletal remains of 37 individuals were analysed; eight moments of burial, from two archaeological sites (Toluquilla and Ranas); from Toluquilla in two buildings (24-27); and seven burials (24-17, 24-20, 24-24, 27-13, 27-13b, 27-14, 27-25). These were adults (46%), infants (43%) and 11% of unborn (see Fig. 3).

Picture 1. Ball court 1 in Toluquilla

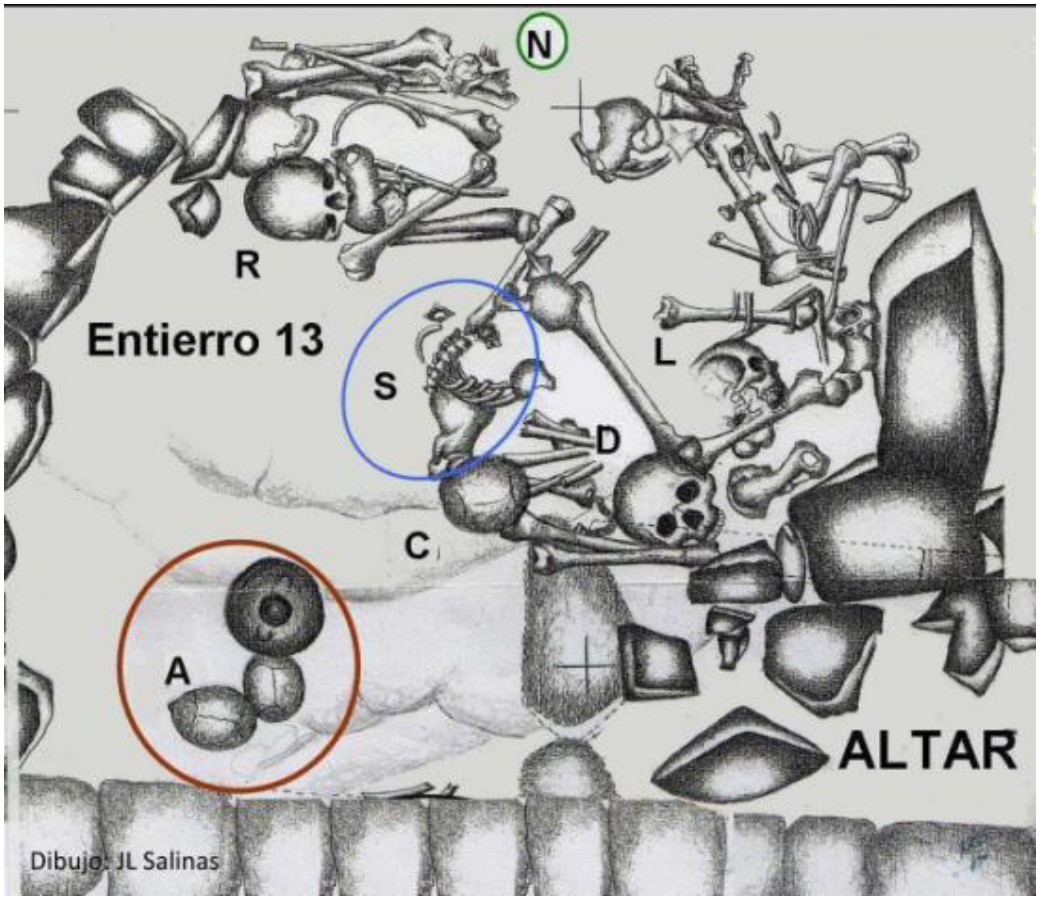

Fig. 2. Burials in building 27, Toluquilla

At the Fig. 4, orange bars represent fetuses; infant's cases are graphed in green and blue adults. All cases of fetuses analyzed everyone exceed 10 mg/kg, and one 23 mg/kg values.

From the toxicological point of view, the inorganic mercury, focuses on clinical cases, is associated with population directly related to mining activities and by exposure to methylmercury in the environment and food. In addition current studies focus on live population, from serologic samples of bodily fluids and there are no data or reference values for the accumulation in the skeletal system.

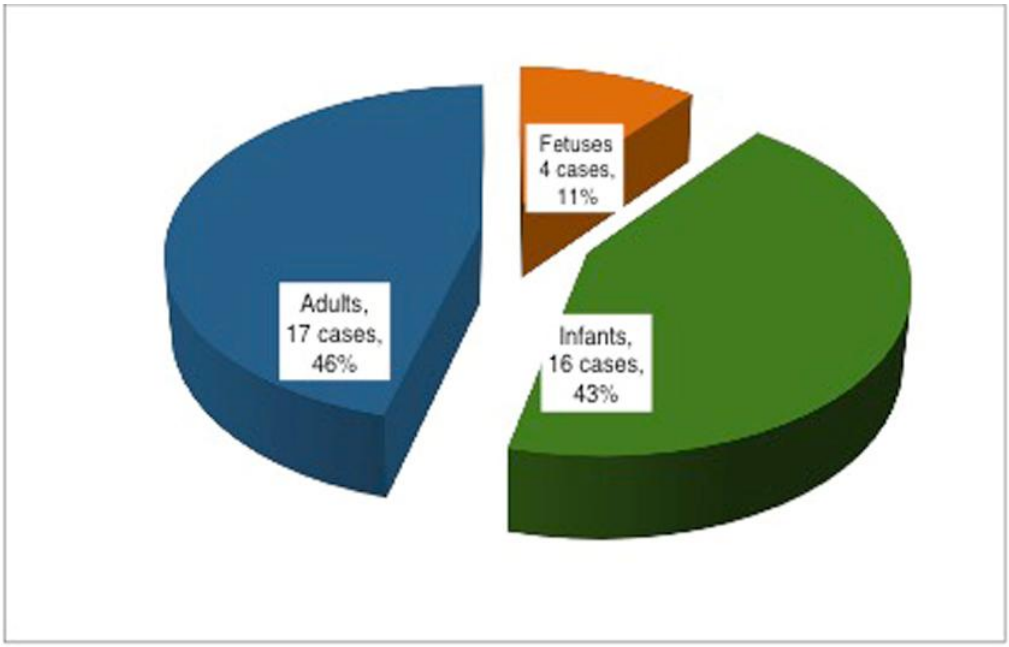

Fig. 3. Pie graph with individual sample composition

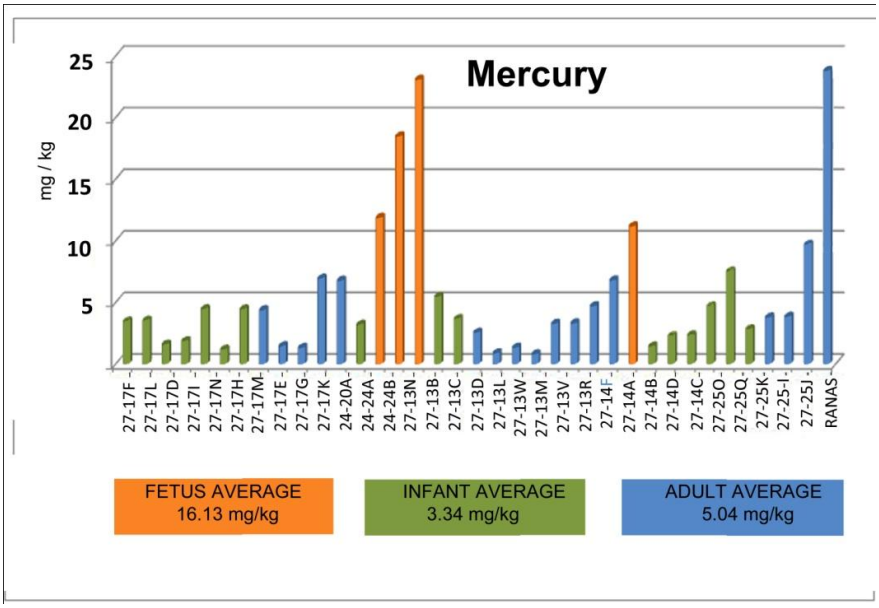

Fig. 4. Measurements of total Hg in bone sample. X axis sets the individual case. Y axis shows the measure of total Hg

Where archaeological, outside of the Sierra Gorda, there are no cases reported for this reason our results have no precedent. In our sample population there are skeletons that have deposits to the inside of the teeth, between dentine, thus mercury is not due to contamination of sediment surrounding him, since it is a product of intake, this is bioaccumulation in life and due to chronic exposure.

Apparently it's a bioaccumulation of alkyl-Hg (methylmercury), that via passing through the placenta are accumulated until the bones of the fetus suffered from symptoms such as a severe infection, as happened in the case of individual F burial 11, although in the case of the individual of burial 24 H did not show apparent effects on the bones but it is possible that its clinical manifestations were in internal organs like liver, brain or kidney.

A pregnant women, by their condition is highly receptive to pollution, which is transmitted to the fetus, to cause harmful effects accelerated as the pregnancy progressed and if she maintained contact with the contaminated dust of mercury and its compounds, they should cause symptoms at the level of soft organs of storage and ultimately the bones.

Therefore, it is not difficult to assume that perhaps the mother died shortly after giving birth, condition that probably could happen on a recurring basis in ancient times. A case of double killing of both the fetus and mother caused by the effect of mercury is difficult to prove since we found the fetus as individual burials.

In this case, we believe only, we can prove how an economic activity of the ancient period can be considered with consequences on the health of the entire population of their time, being more susceptible in our study are pregnant women and children - both fetuses and children under 3 years old-.

The 4 unborn stand out because they have the highest values in all metals within this population, with values of up to 35.20 ppm arsenic and 23.05 mercury, when it should not exceed 3 ppm arsenic and 1.5 ppm of mercury [Mexican standard law is stricter for mercury to set 0.2 parts per million, in the arsenic standard establishes one concentration not greater than 5 parts per million, SEMARNAT, 1993 [17] and [18]. Walters, 1991: 1593 – 1605 [19].

6. FINAL CONSIDERATIONS

The bones of fetuses, have the highest concentration of mercury, which we conclude that it is impacting through the placenta. So they were pregnant women subjected to acute and chronic pollution, which affects the fetus, and sick it. Possibly they were contaminated by breathing cinnabar powder to grind the rocks that contains it, or by eating contaminated meals that affected fetuses health. From this it is concluded that women also participate in the work of mining, milling, or packaging of cinnabar, it is not a task only of males, as proposed by the researchers in the late 1960s (see Picture 2).

Picture 2. Mineral deposit in the teeth of the individual T, in building 27

Whereas children older than two years after ablation are contaminated by intake in some way, or the breath of powders with mercury content. Therefore pregnant women, fetuses and children are a vulnerable population. Minors were not direct labor pollution product, consider conditions where mercury was a public health problem that affected the entire population and the ecological system by i-Hg to alkyl-Hg that lived in pre-Hispanic times in the South of the Sierra Gorda.

The results of this study provide evidence of bioaccumulation from mercury exposure, particularly in the fetus from maternal exposure. Studies consistently show that fetal cord blood mercury levels are higher than the maternal blood levels (Nyland et al. [20], Lederman et al. [21]). The findings from this current study provide further evidence of the vulnerability of the fetus showing the actual mercury accumulation in the fetus. Although this is ancient data, it has implications today since mothers continue to be exposed to mercury from various sources.

7. CONCLUSION

This work is an example how prehistory mining of mercury and their inherent activities are factors of environmental degradation and population health problems since ancient times.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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