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Bone cadmium and lead in prehistoric inhabitants and domestic animals from Gran Canaria

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Abstract

Both lead and cadmium exposures derive from natural sources and also from industrialisation and certain habits, such as cigarette smoking in the case of cadmium. Some of these sources only affect human beings. The aim of this study was to determine the levels of lead and cadmium in bone samples of 16 prehispanic inhabitants of Gran Canaria, 24 prehispanic domestic animals (sheep, goat and pigs) from this island, 8 modern individuals, and 13 modern domestic animals. We found that modern individuals showed higher bone Cd values (mean = 516.7 ± 352.49 μg/kg, range = 167.20–1125 μg/kg) than prehistoric ones (mean = 85.13 ± 128.96 μg/kg, range = 2.97–433 μg/kg). Values of prehistoric individuals did not differ from those of the prehistoric animals (mean = 70.54 ± 46.86 μg/kg, range = 11.06–216.50 μg/kg), but were higher than those of the modern animals (mean = 7.31 ± 10.35 μg/kg, range = 0–35.62 μg/kg). In the same way, modern individuals and modern animals showed approximately 7-fold higher bone Pb than ancient individuals and ancient animals, respectively. Ancient animals showed significantly lower Pb values than all the other groups, whereas modern animals showed Pb values comparable to those of the ancient individuals. A significant correlation was observed between bone Pb and Cd ($r = 0.61$, $P < 0.001$). Since bone cadmium accumulation leads to osteoporosis, we have also tested the relationship between histomorphometrically assessed trabecular bone mass and bone cadmium both in modern and ancient individuals. No significant relationship was found between these two parameters.

Keywords: Bone cadmium; Bone lead; Ancient bones; Canary Islands

1. Introduction

Exposure to heavy metals has been considered to be an epidemic of the Industrial Era. It is assumed that daily absorption of lead by modern American individuals is 100-fold greater (≈29 000 ng) than that of prehistoric ones (Settle and Patterson, 1980). Fumes from car exhausts constitute the main sources of lead (Smith, 1976), so the air in the major cities contains 500–10 000 ng of lead per cubic meter. Therefore, the prohibition of leaded petrol and other measures directed against lead contamination, such as the withdrawal of food

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cans soldered with lead (Settle and Patterson, 1980) has resulted in a decrease in bone lead in many European inhabitants over the last few years compared with the situation some decades ago. On the contrary, bone cadmium levels are rising in Europe (Jaworowski et al., 1985), although bone is not the main organ in which this metal accumulates (Underwood, 1977). Cadmium exposure mainly depends on tobacco smoking but also on industrial pollution (extensive use of cadmium for anti-corrosion plating of metals, construction of batteries, glass and porcelain colouring, photography, lithography, manufacture of silver alloys, etc.) and long-term, low-level exposure from food products may also be important. Chronic cadmium poisoning may be associated with hypertension, proteinuria and tubular dysfunction, emphysema, and osteomalacia and osteoporosis (Nogawa et al., 1975).

In a previous report we showed that bone levels of lead in the prehistoric inhabitants of Gran Canaria were low, although not as low as in other parts of the ancient world, such as Peru, perhaps due to some degree of air contamination derived from intensive mining and the lead and silver industries during the first centuries of this era in Southern Spain, which could have also affected the Canary Archipelago due to the trade winds (González-Reimers et al., 1999). Both lead and cadmium exposure may derive from natural sources and from synthetic materials such as glazed pottery, or, especially in the case of cadmium, from certain habits such as tobacco smoking. Obviously, some of these sources will only affect human beings and not animals. It is, therefore, useful to perform bone analysis both in human and animal remains in order to discern possible sources of exposure to these metals. In order to increase our knowledge about toxic pollutants in the past (and in the present), and to look for differences in bone lead and cadmium between human beings and animals, in this study we report the values of bone cadmium and lead in a prehispanic sample from Gran Canaria, comparing them with those obtained in bone samples of contemporary inhabitants of the Canary Islands. We have also determined bone lead and cadmium in prehistoric domestic animals with an antiquity of 900 years, and compared the values with those of modern domestic animals from the Gran Canaria central highlands that theoretically have low pollution levels. Since cadmium is possibly involved in the development of osteoporosis and osteomalacia, we have also evaluated the relationship between bone cadmium with trabecular bone mass (TBM), both in modern and in ancient population groups.

2. Material and methods

2.1. Samples

2.1.1. Prehistoric inhabitants from Gran Canaria

We have obtained a small sample from the proximal tibial epiphysis of 16 prehispanic individuals buried in Guayadeque, an archaeological site located in the eastern part of Gran Canaria, consisting of several collective funerary caves. The tibiae analysed in this study are housed at the Museo Canario (Las Palmas). We do not know the exact antiquity of these tibiae, but there are some dates for other remains from Guayadeque, which have an antiquity ranging 1405 ± 60–1213 ± 60 bp. They were identified as belonging to 10 males and 6 females, after applying the discriminant functions performed by ourselves for the prehispanic population of Gran Canaria (González-Reimers et al., 2000); age-at-death could only be determined in 5 cases; in 4 of them the proximal epiphyseal closure was still evident, and in another case inspection of the pubic symphysis yielded an age-at-death of less than 53 years old (Brooks and Suchey, 1990).

2.1.2. Prehistoric animals

Samples were found at an archaeological site in Agüimes, also in the eastern part of Gran Canaria, and show an antiquity ranging from 920 ± 70 to 750 ± 50 bp. They belong to 9 goats, 7 sheep and 8 pigs. Age-at-death of these animals is unknown, but they are usually killed before the 6th year of life—and even before in the case of the pigs. These animals were consumed by the prehispanic population, and bone fragments were partially cremated. In all the cases the samples consist of

Table 1
Trabecular bone mass in modern and ancient individuals and bone cadmium and lead in the samples analysed

	Bone cadmium (μg/kg)	Bone lead (mg/kg)	Trabecular bone mass (%)
Modern individuals	516.70 ± 352.49	30.53 ± 14.62	24.28 ± 6.48
Ancient individuals	85.13 ± 128.96	4.06 ± 4.63	16.35 ± 4.32
Ancient animals	70.54 ± 46.86	0.34 ± 0.22	–
Modern animals	7.31 ± 10.35	2.29 ± 2.32	–

long bone fragments, including both cortical and trabecular bone.

2.1.3. Modern reference values

They were obtained from bone samples belonging to 8 men aged 30 ± 9.59 years, who underwent surgical operations on the knee which included removal of a small section of bone from the proximal tibial epiphysis.

2.1.4. Modern animals

The sample included a small section of long bones of 13 modern animals (5 goats, 5 sheep and 3 pigs) which grazed free in the central highlands of Gran Canaria, and were killed for human consumption.

2.2. Trace element analysis

Samples from modern individuals and animals were carefully mechanically cleansed using Milli-Q water. All the samples were then dehydrated in a furnace at 100 °C for 24 h, and then weighed and dissolved in 65% HNO_3 (Merck p.a.) and 10% H_2O_2, in order to digest organic material. The digestion solutions were quantitatively transferred to volumetric flasks and diluted to 10 ml with ultrapure water (Milli-Q OM-140 deionization system). Dry weight of the samples was 653.16 ± 536.55 mg (range = 26–2622.2 mg).

Cadmium and lead were measured using a graphite furnace and a Perkin Elmer atomic absorption spectrophotometer. Detection limits for cadmium and lead were 0.05 and 0.02 μg/kg, respectively.

Prior to the analysis of each element we prepared a blank with ultrapure deionised water (Milli-Q system), and different solutions, at known concentrations, using certified standards of 1000 mg/kg for cadmium and lead (Fisher, Fairlawn, NJ). These solutions were used for the calibration of the apparatus. Correlations between the certified values and those determined by the spectrophotometer were nearly linear.

2.2.1. Trabecular bone mass assessment

A small portion of the medial part of the posterior aspect of the proximal epiphysis was removed and processed for undecalcified bone sample analysis. Briefly, samples were embedded in methylmethacrylate (Sigma Chemicals, St. Louis, MO), stored during 24 h at 4 °C, and later polymerised at 32–34 °C for 3–4 days. Embedded samples were then cut into 9–12-mm thick slices with a Reichert-Jung microtome—so that the resulting sections were perpendicular to the long axis of the tibiae—and stained with toluidine blue. TBM was determined using an image analyser equipped with the program 'Image Measure 4.4a' (Microscience Inc.), at 40×. Results are given as % of total area.

3. Results

In Table 1, it can be seen that modern individuals show higher bone Cd values (range = 167.20–1125 μg/kg) than prehistoric ones (range = 2.97–433 μg/kg). Values of prehistoric individuals did not differ from those of the prehistoric animals (range = 11.06–216.50 μg/kg), but were higher than those of the modern animals (range = 0–35.62 μg/kg).

Similarly, modern individuals showed higher bone Pb (range = 5.65–56.61 mg/kg) than the ancient ones (range = 0–14.76 mg/kg). Ancient animals showed significantly lower Pb values than all the other groups (range = 0.08–0.81 mg/kg), whereas modern animals showed Pb values comparable to those of the ancient individuals (range = 0–7.82 mg/kg).

No correlation was observed between bone Cd and TBM; however, a significant correlation was observed between bone Cd and bone Pb ($r = 0.613$, $P < 0.001$), which was present both in the prehistoric sample ($r = 0.54$) and in the modern one ($r = 0.578$). No significant correlations were found between bone Cd and bone Pb and age, either in the prehistoric individuals or in the modern ones.

4. Discussion

Lead and cadmium may be considered to be environmental poisons of the industrial era. Industrial use of cadmium in the US has doubled every decade since 1900 (Kjellström, 1979). Cadmium concentrations in renal cortex show a relentless increase from almost 0 at birth to approximately 30–70 mg/kg at age 50. Indeed, approximately 45% of the total cadmium body burden—which may reach 20–300 mg in a 40–60-year-old adult not exposed to abnormal amounts of cadmium—accumulates in the liver and kidney, and only a relatively small part is stored in bone (Christoffersen et al., 1988). On the contrary, lead is efficiently retained in the calcified tissues, and over 90% of the body burden of lead is located in the skeleton, although lead accumulation may be different in trabecular bone than in cortical bone (Manea-Krichten et al., 1991).

There are few studies that have focused on bone cadmium in ancient populations. Jaworowski et al. (1985) report Cd values ranging 120–470 ppb (μg/kg) in Ancient Romans, between 110 and 260 in Merovingians, between 100 and 420 ppb in middle age, and between 90 and 800 ppb in modern humans (Jaworowski et al., 1985). In Yjengar's compilation (Yjengar et al., 1978), values as high as 4.2 and 1.28 ppm were reported, although Lindh et al. (1980) found values of only 38 ppb in exposed workers, and Knuuttila et al. (1982) reported mean cancellous bone cadmium concentrations of 0.22 ± 9.16 mg/kg. Baranowska et al. (1995) reported values of 0.4–1.5 ppm Cd (wet weight) together with bone lead values ranging 20–200 ppm in modern Silesian subjects. Bryce-Smith et al. (1977) reported values in stillbirth ranging from 0 to 31.5 ppm Cd. However, bone cadmium was below the detection limit in human teeth from Neolithic and Bronze Ages cemeteries from South Poland (Glén-Haduch et al., 1997), and very low concentrations have been reported for circumpulpal dentin of ancient Nubians (14–72 ppb, Grandjean and Jorgensen, 1990) and North American Peco Indians (32 ± 13 ppb, Ericson et al., 1991). Although in most studies cadmium concentrations in calcified tissues are higher in modern population groups than in ancient ones, the aforementioned data suggest that the impact of current environmental cadmium pollution is much less than that of other pollutants such as lead.

In the ancient individuals from Gran Canaria we have observed bone lead concentrations similar to those reported for both the prehispanic population of Gran Canaria (4.41 ± 3.45 mg/kg, González-Reimers et al., 1999) and Tenerife (4.12 ± 4.77 mg/kg, Arnay-de-la-Rosa et al., 1998). Thus, this study confirms that in prehispanic times, bone lead of the population of the Canary Islands was low, although not as low as in other population groups of the ancient world, such as ancient Nubians (0.4–1.5 mg/kg and 1.4–3 mg/kg during the Pharaonic period, Grandjean et al., 1979), ancient Peruvians (0.11–2.7 mg/kg, Ericson et al., 1979; 0.06–1.9 mg/kg, Drasch, 1982), PreColumbian Southwest American Indians (0.04–1.87 mg/kg, Patterson et al., 1991), or in secondary dentin samples of ancient Greenlanders (0.39–0.95 mg/kg, Grandjean and Jorgensen, 1990). There are no lead mines in the Canary Islands, and metal artifacts were unknown for the prehistoric population of the Islands. Moreover, lead content of soil samples is low (González-Reimers et al., 1999). On the basis of these data and the fact that North–East trade winds blow in the Islands especially during the second half of spring and summer, we hypothesized that perhaps some amounts of polluted air derived from intensive silver and lead mining in Southern Spain during the Roman period could have reached the Canary Islands.

The considerably lower values of bone lead concentrations observed in the prehistoric animals may be explained by the fact that animals were probably killed at a maximum age of 6 years (usually, the killing of pigs takes place earlier, and goats and sheep older than 6 years rarely produce milk). Lead accumulates slowly and progressively in bone (Manea-Krichten et al., 1991). In this study, bone lead concentrations of the ancient individuals is approximately 10–14-fold higher than those of ancient animals. As mentioned previously, although age-at-death could only be determined in 5 prehistoric individuals, the mean age-at-death which has been estimated for the ancient population of Gran Canaria ranges 30–40 years, a figure which is probably 8–10-fold greater than that estimated for the ancient animals analysed in this study. It is, therefore, likely that both animals and human beings were exposed to similar amounts of lead. The same is true for the modern animals, which had much lower bone lead values than those of modern humans, although significantly higher than those of the ancient animals. Indeed, lead concentrations in modern animals are approximately 7-fold those observed in the ancient ones, a proportion which is similar to that existing between modern and ancient individuals.

The pattern observed for bone cadmium, however, is different. In sharp contrast with bone lead contents, which show the lowest values both in ancient domestic animals and prehispanic inhabitants, bone cadmium concentrations are lowest in the modern animals, not in the ancient ones. In fact, the values obtained for the modern animals from Gran Canaria are strikingly low. Cigarette smoking is one of the main sources of cadmium exposure. The differences between bone cadmium in modern individuals and modern animals might be explained on this basis—although, unfortunately, the smoking habit of the modern individuals are not known. The fact that the modern animals showed lower bone cadmium values than the ancient ones is difficult to explain. Perhaps there is some degree of soil contamination which affected the bone samples belonging to the ancient animals. In contrast with the skeletal remains of the ancient individuals, who were not interred, but deposited in huge collective burial caves on stony or vegetal layers, in a subdesertic environment, animal remains found in ancient settlements were partially cremated and in direct contact with soil. Therefore, some degree of diagenetic changes cannot be ruled out. Perhaps these post-mortem changes account for cadmium values observed in the ancient animals, although the values obtained in this study are of the same order of magnitude as those reported by other authors (Jaworowski et al., 1985), and bone lead values do not support an important role for diagenetic changes.

Overall, bone cadmium values observed in the prehispanic inhabitants are lower than those reported for other ancient populations of the ancient world, except for those cases of bone Cd below the detection limit, such as those reported by Glén-Haduch et al. (1997). It is important to bear in mind that the individuals analysed by these authors belong to the Neolithic and Bronze Age, i.e. they are far older than the prehispanic Canarians, and Ancient Romans, Merovingians, etc. Grandjean and Jorgensen (1990) reported cadmium concentrations in circumpulpal dentin belonging to a medieval Denmark population similar to those reported in this study for the prehistoric population of Gran Canaria. Merovingians, Ancient Romans, and Medieval Danish individuals all lived in a continent, with a long tradition in metal mining and smelting activities, which could lead to some degree of cadmium pollution. Ancient Greenlanders show higher teeth cadmium concentrations—but lower lead ones—which were attributed to dietary habits.

As mentioned previously, and in accordance with previous results, the ancient inhabitants of Gran Canaria showed low—but not negligible—amounts of lead and cadmium. Some amounts of cadmium and lead did contaminate air, water or food since at least the Roman period, and also the ancient Canarian population. It is worth noting that zinc sulphide containing variable amounts of cadmium sulphide is closely associated with lead sulphide in Southern Spain, so there is a possibility that some cadmium reached the atmosphere during smelting of lead minerals. Analysis of the 600, 700, 770 and 800 hectopascal (hPa) levels back-trajectories shows that air masses from different origins, including Southern Europe, arrive at the Canary Archipelago (Sancho et al., 1992), at least in modern times. Moreover, preliminary results of studies dealing with metal concentrations in airborne particulate matter at the Izaña observatory in Tenerife (Montelongo et al., 1991) suggest that different air masses carry different amounts of trace elements: whereas Al, Fe, Mn, Ca, Mg, Co and Ni are components of the Saharan air masses, Zn, Cd, Cu and Pb have quite a different origin, including a Southern European one. Bone lead in ancient individuals and animals suggest a common exposure to the same source of lead, whereas the higher bone cadmium concentrations relative to age-at-death observed in the ancient animals compared with the ancient human beings can be explained by some degree of diagenetic contamination. Interestingly, we have observed a significant correlation between bone lead and cadmium, both in ancient and in modern bones. Although circulation of air masses may have changed over the last few centuries, the results in the ancient bones might support the aforementioned hypothesis of air pollution due to intensive mining in Southern Spain during the Roman period. The results for both lead and cadmium in the modern bones are probably due to industrial pollutants.

We have reported a high prevalence of osteopenia among the prehispanic population of Gran Canaria (Velasco-Vázquez et al., 1999). Some authors have found a relationship between cadmium exposure and decreased bone mass (Jarup et al., 1998), an effect derived from impaired vitamin D synthesis due to kidney damage, together with a cadmium mediated increase in urinary calcium excretion (Staessen et al., 1999) and a direct effect on osteoblast and osteoclast function (Berglund et al., 2000), and, perhaps, on the formation and properties of hydroxyapatite (Blumenthal et al., 1995). The data derived from this study do not support such a relationship, since no correlation was observed between bone cadmium and TBM either in the ancient individuals or in the modern ones.

In conclusion, we found that ancient individuals showed bone lead and cadmium contents approximately 7-fold lower than the modern ones, a proportion similar to that observed when bone lead contents are compared between ancient and modern animals. Bone lead values, both in ancient and modern individuals and animals, point to the existence of a common source of exposure shared both by animals and human beings. In accordance with previous reports, bone lead levels in ancient individuals, although low, are higher than those observed in other studies dealing with ancient population groups, whereas bone cadmium levels of the ancient inhabitants of Gran Canaria were low, in the range of those observed in other ancient population groups. No correlation was observed between bone cadmium and TBM, a result which does not support the existence of a link between osteoporosis and cadmium exposure.

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