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Sensitivity of two biomarkers for biomonitoring exposure to fluoride in children and women: A study in a volcanic area

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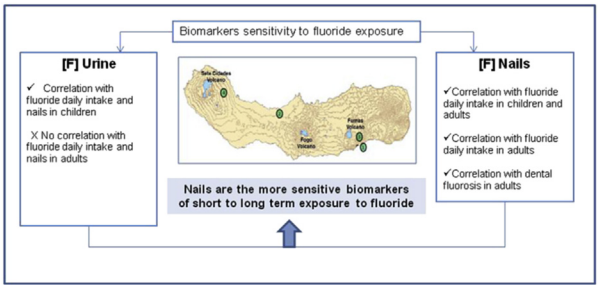
HIGHLIGHTS

- Urine is a viable biomarker for fluoride exposure in children.
- Nail clippings is a useful biomarker for fluoride exposure in children and adults.
- Fluoride daily intake correlates positively with dental fluorosis in adults.
- Nail is a more reliable biomarker of chronic exposure to fluoride than urine.

GRAPHICAL ABSTRACT



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ABSTRACT

The natural enrichment of water with fluoride is related to natural sources such as volcanic activity, with it being documented that fluorosis, an endemic and widespread disease in volcanic areas, is associated to the ingestion of high levels of fluoride through water. Thus, in this study, we aimed to define the fluoride concentration in drinking waters of volcanic origin and compare the sensitivity of urine and nail clippings as biomarkers for fluoride exposure in adults and children.

Samples of drinking water from four villages in São Miguel Island (Azores) were used and the fluoride concentration was determined, as well the fluoride content in urine and toenails clippings from 66 children and 63 adults from these villages. A validated diet questionnaire, assessing sources of fluoride, was recorded for each participant. The fluoride determination in urine and nail clipping samples was made using a fluoride-specific electrode.

A positive correlation was found between the fluoride daily intake and fluoride content in children urine ($r_s = 0.475$; $p < 0.001$) and in their nail clippings ($r_s = 0.475$; $p < 0.001$), while in adult women, the fluoride daily intake correlated positively with fluoride content nail clippings ($r_s = 0.495$, $p < 0.001$).

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This reveals that nail clippings are more reliable as biomarkers of chronic exposure to fluoride than urine for populations of different ages (children vs. adults). Furthermore, nail clippings are more suitable than urine fluoride levels to assess long term exposure to fluoride in areas where the exposure to fluoride in drinking water is considered within, or slightly above, the recommended legal values.

1. Introduction

Fluoride is not an essential element for human growth, but it is incorporated in the mineral part of the bones and in the teeth (WHO, 2002); small amounts, in the order of 1 mg/L in ingested water, have a beneficial effect on the rate of occurrence of dental caries, particularly among children (WHO, 1997). On the other hand, excessive intake of fluoride results in pathological changes in teeth and bones, such as dental and skeletal fluorosis (WHO, 1984).

Fluorosis is a widespread disease and endemic in several regions of the world (Ayoob and Gupta, 2006), being usually related to ingestion of high levels of fluoride, especially through water. Although fluoride content in drinking water is known as an important factor to be controlled in order to prevent adverse health effects, Edmunds and Smedley (2004) estimate that there are more than 200 million people in the world with drinking water that exceeds the WHO maximum level (1.5 mg/L) (WHO, 2004).

The high levels of fluoride content in groundwater are generally related to natural sources, and the association of fluoride with areas of volcanic activity is well documented (D'Alessandro, 2006; Notcutt and Davies, 1999). The fluoride content in volcanic aquifers may result from rock leaching processes and from the rising volcanic gases which are dissolved in the groundwaters (Aiuppa et al., 2003; Cronin et al., 2003). This is particularly important in the Azores archipelago (Portugal), where due to their origin, seismicity and volcanism are frequent in the archipelago, resulting in the existence of aquifers formed by the volcanic rocks that erupted during the principal building stage of each volcano. According to Cordeiro et al. (2012), about 98% of the Azores water supply originates in groundwater sources. In São Miguel Island, the direct effects of volcanogenic fluoride on human populations are well recorded for the village of Ribeira Quente, located only 5 km from the caldera of Furnas volcano, where most of the inhabitants have visible evidence of dental fluorosis in permanent teeth due to raised fluoride levels in the drinking water (Baxter et al., 1999).

Nowadays, high fluoride content in drinking water poses as a public health problem (Mehta, 2013) and there has been an increase in the search of biomarkers for monitoring fluoride intake. The monitoring of human exposure is accomplished by the analysis of several biological tissues or fluids, e.g. teeth, bone, nail, hair, plasma, urine and saliva (Whitford, 2005); however, urine and nails due to their easy collection are the most commonly used matrices.

Urine is recognized as a biomarker that reflects a very recent exposure, during the last hours, to fluoride (WHO, 1994), although urinary fluoride concentrations do not provide a direct measure of fluoride excreted via urine because of variations in urinary flow and pH (Whitford, 1996). On the other hand, current reports suggest that fluoride concentration in fingernail clippings reflects the chronic level of exposure to fluoride (Czarnowski and Krechniak, 1990; Feskanich et al., 1998). The major advantage of using fingernail clippings is that the concentration of fluoride reflects the average level of intake and the plasma concentration over a protracted period, usually 1–2 weeks depending on how often the nails are clipped (Whitford, 2005). Although these biomarkers have been reported for the assessment of fluoride exposure through drinking water, epidemiological studies for fluoride exposure in areas with endemic fluorosis are scarce. To our knowledge, there aren't any studies carried out with both biomarkers to compare their suitability as biomarkers of exposure to fluoride for populations of different ages (children vs. adults) and, in areas where the exposure to fluoride in drinking water is considered within or slightly above the recommended legal values. Thus, in areas where endemic fluorosis has been reported, such as the Azores, and were the fluoride content in the drinking water is in general within the normal range, these studies assume a particular importance. Therefore, the present study aims to investigate whether urine and nail clippings have a higher sensitivity or utility for biomonitoring human population from different age classes that are chronically exposed to fluoride by: i) determining the fluoride concentration in drinking waters with volcanic origin; ii) testing the association between fluoride concentration in nail clippings and urine and, iii) testing the association between fluoride content in these biomarkers with the development of dental fluorosis in humans.

2. Material and methods

2.1. Study areas

The Azores archipelago comprises nine volcanic inhabited islands (Fig. 1A). São Miguel Island, the largest of the archipelago, is formed by three major active central volcanoes, Sete Cidades, Fogo and Furnas (Fig. 1B), connected by a regional lineament of basaltic volcanism. In this volcanic island, the groundwater geochemistry is influenced by the dissolution of primary minerals of the volcanic rocks that occurs in two major aquifer systems (Cruz, 2003): (1) the basal aquifer system, which corresponds to fresh-water lenses floating on underlying salt water, and (2) in perched-water bodies (Cruz et al., 2009).

The selected studied areas are two fishery villages, Ribeira Quente and Porto Formoso, and two rural villages, Furnas and Sete Cidades, located inside the crater of a volcano (Fig. 1B). In the villages of Porto Formoso (PF) and Sete Cidades (SC) the water supply results from groundwater bodies, in the western sector of the São Miguel Island, associated with two active central trachytic volcanoes (Sete Cidades and Fogo) (Cruz and Amaral, 2004); in the villages of Ribeira Quente (RQ) and Furnas (FR), the water supply results from groundwater bodies with origin in various springs associated with the Furnas active volcano.

2.2. Study population

The population selected for this study includes female schoolchildren and their mothers, from a possible match of 105 girls and 102 mothers. A total of 66 children and 63 adult women participated in the study, reflecting a participation rate higher than 50%. Their participation was anonymous and voluntary. Adults were only considered eligible to participate in the study if they were “stay-at-home” mothers between the ages of 25 and 50. Children between the ages of 4 and 12 were considered. Both the children and the adults that participated in this study were born and raised