FLUORIDE ACCUMULATION IN HUMAN SKULLS 
IN RELATION TO CHRONOLOGICAL AGE

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SUMMARY: Fluoride concentration in 200 bones of human crania were determined by fluoroselective electrode. The fluoride content increased with the prolongation of the chronological age of examined skulls. It was found that the significant (p < 0.001) bone fluoride enriching takes at least around 1000 years. Sex is not a factor differentiating the fluoride content of skulls.

Key words: Bone fluoride; Fluoride content; Human skulls.

Introduction

Fluoride is present in all tissues and fluids of animal organisms, with the highest concentrations occurring in bones and teeth.1 It accumulates steadily in bones, and the content increases with age.2 As early as in fetogenesis a slow but regular increase of fluoride concentration in the skeleton is observed.3,4 Much higher concentrations of fluoride, the result of adsorption from the surroundings, have been found in bones of fossil animals in comparison with skeletons of modern ones.5 Thus, fluoride tends to be incorporated into bone structures, both during life and after death. These observations prompted us to carry out some studies on large collections of archaeological material, to seek an answer to the question of correlations between bone fluoride content and chronological age.

Materials and Methods

The research material consisted of 200 human skulls (86 female and 114 male) from collections of the Department of Anthropology of the University of Wroclaw and the Department of Anatomy of the Pomeranian Medical Academy in Szczecin.

Bone powder was sampled, with use of a dental drill, from the area of the external occipital protuberance, after previous surface cleaning with removal of a thin superficial layer of bone. The material was solely from the bone cortex. To determine the fluoride content of the samples, 20 mg were first dissolved in 0.3 mL of 2.0 M HClO 4, then neutralized in 1.2 mL of 0.5 M solution of trisodium citrate, and finally 0.5 mL of TISAB buffer (pH = 5.5) was added. The level of fluoride was determined with the Orion-96-09 fluoroselective electrode (Orion). The crania were examined by an anthropologist to establish chronological age and sex. The results were statistically analysed by the Kolmogorov-Smirnov test at significance level p < 0.001.

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### TABLE

Fluoride content (ppm) of examined skulls in relation to the locality of excavation, age and sex

<table>
<thead>
<tr>
<th>Locality of excavation</th>
<th>Age</th>
<th>Female No. of cases</th>
<th>Range</th>
<th>Male No. of cases</th>
<th>Range</th>
<th>Total No. of cases</th>
<th>Mean (x)</th>
<th>SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Złota</td>
<td>4000-1700 BC</td>
<td>11</td>
<td>647.3-1756.5</td>
<td>11</td>
<td>687.5-1929.7</td>
<td>22</td>
<td>1105.8</td>
<td>363.4</td>
</tr>
<tr>
<td>Plonsk</td>
<td>10-12th AD</td>
<td>13</td>
<td>144.2-458.9</td>
<td>13</td>
<td>149.0-418.9</td>
<td>26</td>
<td>285.5</td>
<td>88.3</td>
</tr>
<tr>
<td>Sandomierz</td>
<td>10-12th AD</td>
<td>11</td>
<td>155.8-372.4</td>
<td>11</td>
<td>173.4-451.9</td>
<td>22</td>
<td>273.7</td>
<td>82.9</td>
</tr>
<tr>
<td>Głogów</td>
<td>10-14th AD</td>
<td>12</td>
<td>100.6-342.3</td>
<td>13</td>
<td>102.1-343.1</td>
<td>25</td>
<td>226.3</td>
<td>75.9</td>
</tr>
<tr>
<td>Czeladź W.</td>
<td>12-14th AD</td>
<td>7</td>
<td>107.4-297.5</td>
<td>5</td>
<td>190.7-273.3</td>
<td>12</td>
<td>215.7</td>
<td>50.6</td>
</tr>
<tr>
<td>Sypniewo</td>
<td>13-15th AD</td>
<td>12</td>
<td>93.7-257.6</td>
<td>12</td>
<td>130.9-272.3</td>
<td>24</td>
<td>182.1</td>
<td>47.7</td>
</tr>
<tr>
<td>Wrocław</td>
<td>15-16th AD</td>
<td>11</td>
<td>102.6-278.9</td>
<td>9</td>
<td>139.8-217.0</td>
<td>20</td>
<td>175.8</td>
<td>46.7</td>
</tr>
<tr>
<td>Lublin</td>
<td>18-19th AD</td>
<td>9</td>
<td>113.7-196.6</td>
<td>10</td>
<td>110.4-199.6</td>
<td>19</td>
<td>159.7</td>
<td>31.6</td>
</tr>
<tr>
<td>Szczecin</td>
<td>20th AD</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>30</td>
<td>165.5</td>
<td>58.1</td>
</tr>
</tbody>
</table>
These procedures included measurement of the average fluoride concentration of skulls of different chronological age from eight exploration sites in Poland (Zlota, Plonsk, Sandomierz, Glogow, Czeladz Wielka, Sypniewo, Wroclaw, and Lublin - see Map). The modern skulls from Szczecin composed the control group.

Results and Discussion

Our results (Table) show that fluoride content of skulls increases with chronological age, and that significant (p < 0.001) bone fluoride enriching takes at least around 1000 years.

Higher fluoride concentrations were found in male skulls, except those from Plonsk, but the sex differences were not statistically significant. We therefore conclude that sex is not a factor differentiating fluoride content of skulls.

The oldest crania (from Zlota) have on average 4 to 7 times higher fluoride content when compared with the other groups and the control group. Comparison of fluoride content in the medieval skulls with the control group showed significant statistical differences in the case of skulls from Plonsk and Sandomierz. If we accept that the majority of skulls from Plonsk and Sandomierz are dated back to the 10th, and not to the 12th century, the time span dividing them from the control group skulls would be around 1000 years. Given that assumption, one could propose that significant bone fluoride enrichment takes at least around 1000 years. Taking into account the facts that the particular regions of Poland where the skulls come from do not differ significantly in climate, and that the soil fluoride content ranges from 80 to 550 ppm in Poland, we find the time factor to be the main cause of the observed differences. This finding is confirmed by comparison of the fluoride content of skulls from Zlota (4000 - 1700 BC) and Sandomierz (10th - 12th century). Both sites are very close to each other, and both are located on loesses having similar fluoride content, yet the average fluoride content of skulls from Zlota is four times higher than that in skulls from Sandomierz. Only this clear difference in chronological age can explain such significant differences in fluoride content.

In 1844 Middleton stated that the content of fluoride in fossil bones increases with chronological age. Oakley and Straus are of the opinion that the greater part of fluoride in fossil bones is built into them after death. In 1992 Gonzalez-Reimers and Arnay-de-la-Rosa presented results of studies on seven human skeletons from the Canary Islands, dating back to the prehispanic period. The concentration of fluoride in these bones was 10 times greater than that in the soil, which could be indirect evidence of fluoride transmission from soil into bones over time. Parker and Toots, on the other hand, thought that though the trend of bone fluoride content increasing is universal, the intensity of the process depends largely on the accessibility of fluoride in the given area. By use of electron microprobe analysis the authors included fluorine in a group of microelements that enter the structure of apatites as a result of post-mortem processes related to the accessibility of this element in the surroundings, to time and, to a lesser degree, to the environmental conditions. Temperature was mentioned as one of the conditions, and its rise favours the substitution of hydroxyl ions with fluoride. Fossil bones coming from tropical zones had significantly higher fluoride content than bones of comparable age excavated in temperate zones.
Conclusions

1. The fluoride content of human skulls increases with their chronological age. The evidence is the accumulation of bone fluoride, in particular after death within the time span from burial to excavation.
2. This accumulation is a slow process, and significant bone fluoride enrichment takes at least around 1000 years.
3. Sex is not a factor which significantly influences the fluoride content of skulls.

Acknowledgement

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