Lead content of deciduous tooth enamel from a high-radon area

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Lead concentrations in the enamel of deciduous incisors of 49 6- to 7-year-old children living in Askola, a rural area in which the radon level is one of the highest in Finland, were determined by the proton-induced X-ray emission method. The absolute concentrations were obtained by calibration with the animal bone standard of the International Atomic Energy Agency. The mean lead concentration of 8.8 ± 6.6 ppm of the whole enamel agreed well with the earlier corresponding lead data from other regions of Finland, indicating that no significant increase in the lead level of the teeth would have occurred because of radon decay. However, the lead concentration level measured on the tooth surface was somewhat higher in Askola, 232 ± 141 ppm, than in the low-radon area Oulu (167 ± 139 ppm; $p < 0.10$). The lead concentration of the whole enamel of the upper incisors, 12.4 ± 8.0 ppm, was twice as high as that of the lower incisors, 6.8 ± 4.6 ppm ($p < 0.005$), emphasizing the importance of classifying lead concentration data by tooth type. ☐ *Children; PIXE; radiation exposure*

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Radon (Rn) exposure is known to be an uncontested health threat; for example, it increases noticeably the incidence of lung cancer (1). Radon is a radioactive gas produced through the decay of radium-226, a naturally occurring element present in trace amounts throughout the earth's crust. Of the natural radiation exposure in Finland at least 40% is due to the decay products of the ^{222}Rn isotope (2). One way to study the cumulative effect of radon in man is to measure the decay rates of the long-lived daughters of ^{222}Rn—that is, those of ^{210}Pb and ^{210}Po in teeth. However, because of the low concentrations of these isotopes the decay yields are low and cause large error margins both in the radiation activity and concentration determinations of these isotopes (3, 4). An easier, although not quite conclusive, method of measuring the cumulative effect of radon would be to measure total lead in teeth, since, in addition to the radioactive isotope ^{210}Pb, a stable product of the ^{222}Rn chain is ^{206}Pb.

Although the increase in the lead content of air due to radon decay is normally insignificant—for example, the radon activity of 10,000 Bq produces 0.3 pg lead per day—the significant lead increase in other ambience such as soil, water, and dust is evident owing to the long exposure time. In accordance with the lead content measurements of soil (5) and the radioactivity measurements (6) in Finland, the high lead values occur in the regions with high radioactivities. Especially, the intake of house dust contaminated with lead increases significantly the levels of lead in children (7). On the basis of these assumptions the main purpose in the present work was to test whether there is any correlation between the lead content of teeth and ambient radon activity. For this purpose the lead concentrations of deciduous incisors collected from children living in the rural community of Askola, situated in one of the regions with the highest natural radioactivity, and in the small town Oulu, in the low-radioactivity region in Finland, were determined (6). The median value of radon activity measured in room air in Askola is

540 Bq/m^3 (maximum value, 56,000 Bq/m^3), whereas for the whole country it is 90 Bq/m^3 (6). In this region the radon content of bored well water ranged from 500 to 55,000 Bq/l.

Materials and methods

Teeth were collected in 1984–85 from two Finnish communities, Askola, a rural, non-industrialized, and high-radon area, and Oulu, an urban, industrialized, and low-radon area (6). The teeth collected were exfoliated upper and lower incisors, and the donors' age was 6–7 years. All teeth were analyzed individually, and whenever one child contributed more than one tooth, they were analyzed as one unit; however, lower and upper incisors were analyzed separately. All teeth were free of visible caries or defects.

All teeth were cleaned with a rotating brush and distilled water and dried finally with an air spray. For the preparation of pellets teeth were dried overnight at 105°C and then ground to a fine powder in a mechanical mill; enamel and dentin were separated by the flotation method (8). The enamel powder was compressed into pellets, 5 mm in diameter and 0.5–1 mm thick. The pressing faces were coated with titanium nitride so that no contamination occurred.

The PIXE method was used for detection of lead. The proton beam was generated with the 2.5-MV van de Graaff accelerator at the University of Helsinki, and X-rays were detected with a 50-mm^2 × 6-mm intrinsic germanium detector, which had an energy resolution of 180 eV for the FeK$_\alpha$ peak during data collection. The X-ray spectra recorded on 1024 channels were stored in the 4K memory of a PDP-9 computer. The main features of the experimental set-up in this study are almost the same as in the work of Anttila et al. (9). For the measurements when the samples were in pellet form the beam sweep system was used, and the beam spot on the sample was approximately 7 mm^2. For the direct surface enamel measurements the smaller spot of 1 mm^2 was used owing to the small size of the deciduous tooth samples. Because the surface smoothness and the sample-detector distances varied in the direct surface measurements, the Pb values were normalized to a calcium content of 37.2%, the mean value found in the powder pellet measurements.

The lead concentration values were calculated by comparing the intensity of the lead L$_\alpha$ peaks of the sample with those of the IAEA animal bone (H-5) standard (10), for which a lead concentration value of 2.97 (SD, 0.26) ppm is reported. The main composition of the IAEA bone standard is so close to that of the tooth that no correction is necessary to compensate for the stopping power of protons. In addition, the calcium concentration value was checked against a $Ca_{10}(PO_4)_6(OH)_2$ sample. With measuring periods of 10 min the detection limit for lead was about 0.5 ppm.

Student's *t* test was used to test the significance of the differences in the mean lead concentrations.

Results

The results of the present Pb concentration measurements in the enamel of deciduous incisors collected from children living in Askola and Oulu are given in Table 1. The mean Pb content of 8.8 ± 6.6 ppm for the whole enamel samples from Askola and the mean values of 232 ± 141 and 167 ± 139 ppm for the surface enamel (at a 0- to 20-μm depth) from Askola and Oulu, respectively, were deduced.

Three typical X-ray spectra taken from the enamel of the same tooth are illustrated in Fig. 1. The bromine peaks occurring as impurities in the pellet spectrum of Fig. 1 originate from the process of separating the enamel and dentin. As shown in Fig. 1, the relative peak heights in the PIXE spectra taken first from the labial and lingual surfaces and then from the powdered enamel sample prepared from the same tooth are clearly different; that is, the relative concentrations of the trace elements vary remarkably.

Table 1. Mean concentrations of lead in the enamel of deciduous incisors from two communities in Finland

Community	Tooth type	Sample type	*n*	Lead concentrations, ppm, mean ± SD		*p*
Askola	Lower incisor	Powder	31	6.8 ± 4.6	8.8 ± 6.6	0.005
	Upper incisor	Powder	18	12.4 ± 8.0		
Oulu	Upper incisor	Labial surface	9	189 ± 211	232 ± 141	0.10
	Upper incisor	Lingual surface	9	276 ± 224		
	Upper incisor	Labial surface	19	244 ± 212	167 ± 139	
	Upper incisor	Lingual surface	19	90 ± 83		

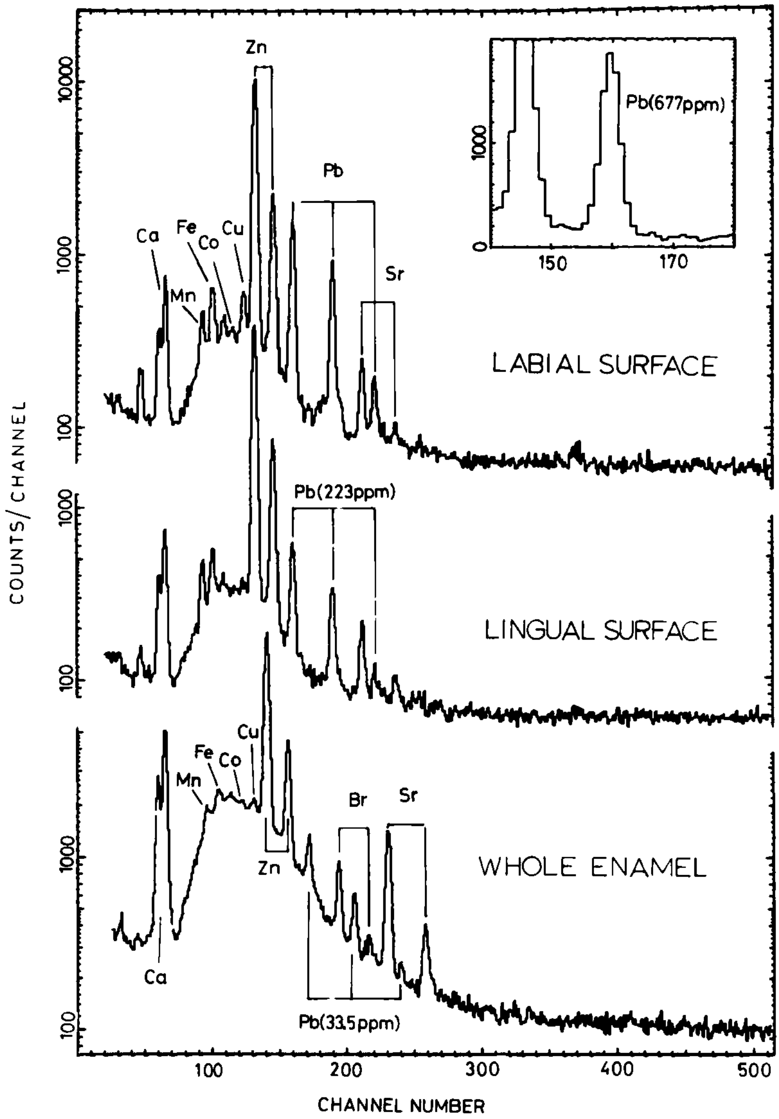

Fig. 1. Comparison of X-ray spectra taken from the same tooth enamel, from the labial surface, from the lingual surface, and from the powdered enamel pellet. A plotting constant of 50 has been added to raise the spectrum from the base line.

Discussion

By means of the proton-induced X-ray emission method (PIXE) lead concentrations in teeth down to 0.5 ppm can be determined in rather short runs (some minutes). The lead concentrations in enamel are seldom below this limit, whereas in dentin such values can occur more frequently (11). As illustrated with the spectra in Fig. 1, the use of PIXE is suitable and recommendable for elemental analysis of tooth surfaces, especially for the analysis of those trace elements marked in Fig. 1.

The thickness of the surface layer from which the Pb concentration with PIXE was detected is 0–20 μm. Owing to the nature of the method the probing efficiency is greatest on the very surface, and it decreases rapidly inwards. Because the lead content in the surface measurements was a factor of approximately 20 higher than the average value for the enamel, the lead had concentrated strongly on the surface. The Pb content of the enamel surface from Askola ranged from 52 to 687 ppm, compared with the corresponding values of 1.2 and 33 ppm in the whole enamel. Thus the lowest value on the surface was higher than the highest one for the whole enamel. Although the average for the lead on the surface in Askola was higher than in Oulu, the highest individual lead value of 860 ppm was measured in a sample from Oulu. On the basis of the present results it seems that there is no clear pattern in the occurrence of lead on the surface. The Pb concentration on the labial surface of the teeth from Oulu was generally higher than that on the lingual surface; however, in the teeth from Askola the inverse was true in many cases (see Table 1 and Fig. 2). To increase the comparability, a scan over the tooth surface should be performed. However, although for the high Pb content on the tooth surface good local accuracy is achieved in a short run (≈1 min), a scan of the whole tooth surface is very difficult.

Only one earlier surface study concerning Pb determinations on the surface of deciduous teeth is available, according to the recent review article of Cutress (12). The reported mean value of 235 ppm for the molars and incisors (13) agrees with the present overall mean values of 232 ± 141 and 167 ± 139 ppm from Askola and Oulu, respectively, for the incisors. However, their mean value of 38 ppm for whole tooth enamel was remarkably higher than the present overall mean value of 8.8 ± 6.6 ppm.

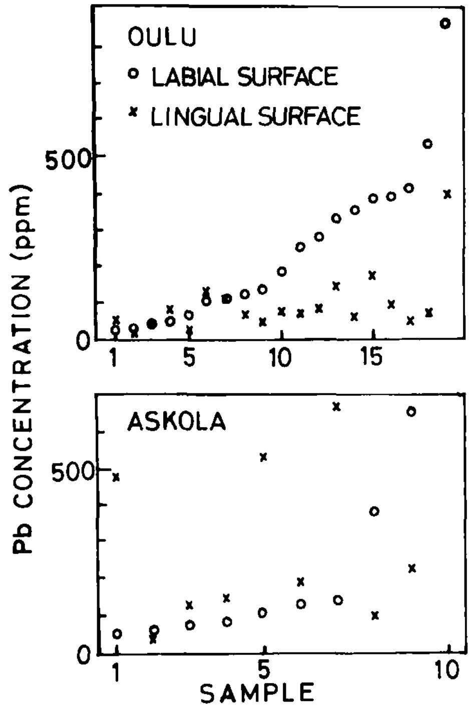

Fig. 2. Coincident presentation of lead concentrations of the labial and lingual enamel surface in the same individual teeth from Oulu and Askola children. The samples are plotted in order of increasing lead concentrations of labial surface.

It has previously been pointed out that the lead content of incisors is the highest and that of molars the lowest (14–17). In the present work the lead content of the upper incisors (12.4 ± 8.0 ppm) was observed to be twice that of the lower incisors (6.8 ± 4.6 ppm). These results would agree with the concept presented by Stack (18) that the lead concentration increases from the posterior to the anterior and from the lower to the upper jaw.

In using lead determinations to examine the cumulative effect of radon, the complicating factor is to decide on the origin of lead. On the other hand, because the cumulative lead exposure of children can be determined by the tooth-lead analysis of deciduous teeth, a finding of high lead concentrations should in any case be investigated further as to the origin of the lead (19). Comparison of the present overall mean lead content with that of 9.0 ± 1.4 ppm derived from the earlier Finnish results (11, 20–22) shows that no clear increase in Pb level due to the radiation has occurred. The lead due to the traffic and industry could be considered to compensate for radon-derived lead in the samples from towns. However, in Helsinki, the largest town in Finland, there was no significant lead accumulation difference due to traffic; according to Pönkä et al. (20), lead concentrations in enamel from the high- and low-traffic areas were 10.6 ± 5.4 and 11.6 ± 3.9 ppm, respectively. Very similar results have been reported by Lockeretz (15). Accordingly, the moderate lead content variations of air would have only a slight effect on the lead content of teeth. However, because the large lead content variations in teeth occur individually, the main part of lead in teeth originates from the other sources such as food, drinking water, dust, or pica. The radon exposure may increase the lead content level of these, as it seems to do in soil (5, 6).

The surface measurements indicate that the Pb level is slightly higher in samples from Askola (232 ± 141 ppm) than in those from Oulu (167 ± 139 ppm) ($p < 0.10$). To confirm definitely that there is relationship between the lead content of the teeth and ambient radon radioactivity, detailed radioactivity measurements should be performed where the lead level of children's teeth is exceptionally high or low.

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