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Aspects of the geographical variations of naturally occurring $^{210}Pb/^{210}Po$ in permanent teeth of juveniles in the UK

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Abstract.

Purpose: To study geographical variations in the level of naturally occurring ^{210}Pb-supported ^{210}Po in permanent teeth of juveniles in the UK.
Materials and methods: Permanent teeth extracted from 278 juveniles for orthodontic purposes were obtained from 48 counties in the UK. ^{210}Po activity concentration was measured on the outer enamel surface using TASTRAK α-particle-sensitive plastic track detectors.
Results: Geometric mean $\pm$ SE activity concentrations in teeth from urban, suburban and rural areas, excluding the high radon area of Devon, were 8.41 $+0.25/-0.24$, 7.76 $+0.37/-0.35$ and 7.20 $+0.49/-0.46$ Bq kg^{-1}, respectively. Overall, there was no significant association between α-activity on the outer enamel surface of permanent teeth and proximity to the major UK motorways. However, when the data were considered with respect to the prevailing south-westerly wind on the western side of the UK, a statistically significant association with respect to donors living downwind (on the easterly side) of the motorways was found. This effect was greater for sections of the M5 and M6 motorways that traverse urban areas. ^{210}Po levels in teeth were also associated with domestic radon concentration. This effect was comparable with that from traffic and urban pollution.
Conclusions: Higher levels of ^{210}Pb-supported ^{210}Po are seen in permanent teeth of juveniles near sources of increased exposure in the UK. Inhalation uptake is an important pathway of exposure, especially with respect to domestic radon exposure. The results might be important in assessing integrated exposure to ^{210}Po in the skeleton and consequent high linear energy transfer dose to bone marrow.

1. Introduction

A previous paper described the morphological features of the microdistribution of natural α-radionuclides in children's teeth (James *et al.* 2004). The most important emitter present is ^{210}Pb-supported ^{210}Po (hereafter for simplicity termed ^{210}Po) and the highest levels of ^{210}Po are found on the outer enamel surface of the tooth. The second most important emitter is ^{226}Ra and this is found preferentially concentrated in the circumpulpal region.

In bone, ^{210}Po is the most important long-lived α-emitter present at typical concentrations in the range 2–9 Bq kg^{-1} (Hunt *et al.* 1963, Hill 1965, Jaworoski 1969, Henshaw *et al.* 1988), whereas average ^{226}Ra concentrations are around 0.25 Bq kg^{-1} (Fissenne *et al.* 1981). In teeth, average ^{226}Ra concentrations are also around 0.25 Bq kg^{-1}, but ^{210}Po concentrations can be much higher. James *et al.* (2004) concluded that the α-activity on the outer enamel surface of teeth best represents a measure of cumulative exposure to environmental ^{210}Po via its ^{210}Pb precursor. Average activity concentrations on this surface were around 10 Bq kg^{-1} with individual values extended to over 30 Bq kg^{-1}.

These observations follow from findings in bone where enhanced deposition on endosteal surfaces has also been observed. Salmon *et al.* (1994) used α-particle spectroscopy in TASTRAK plastic to study ^{210}Po activity on cortical and trabecular surfaces of human femur and cranium. Evidence was found of a fourfold increase in activity concentration in a narrow band at bone surfaces $<3\,\mu$m deep. Salmon *et al.* (1995) used real-time α-spectroscopy to make similar observations in Canadian Arctic caribou, but with much greater precision since these animals contain levels of ^{210}Pb and ^{210}Po 100–1000 times higher than their human counterparts. In the femur, the authors found surface concentrations of ^{210}Po in a layer 1.9–6.4 μm thick, in which activity concentration was enhanced 1.5–10 times with respect to diffuse volume-distributed ^{210}Po. Manea-Krichton *et al.* (1991) and Lovaas and Hursh (1968) reported correlations between stable lead in permanent teeth and that in various adult bones. Overall, the evidence suggests that ^{210}Po on the outer surface of teeth might serve as a marker of levels in bone of relevance to the resultant high-linear energy transfer (LET) dose to bone marrow from this important natural α-emitter.

The present authors previously reported higher ^{210}Po α-activity concentration in the teeth of children living near motorways in the UK (Henshaw *et al.* 1995). This paper reports the results of a targeted analysis of ^{210}Po α-activity in permanent teeth with respect to specific potential sources of elevated

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exposure. These include (1) urban traffic pollution, (2) motorway traffic pollution and (3) domestic radon concentration.

2. Materials and methods

2.1. *Tooth collection*

Permanent teeth extracted from juveniles for orthodontic purposes were obtained from across the UK by arrangement with Local Health Authority District Dental Officers. For each tooth, information on tooth type, the age of the donor, gender and the postcode of the donor's address was requested. No further information was available: in particular there was no provision for asking further questions concerning the donor, either retrospectively or at the time of tooth donation.

Teeth were predominantly of the premolar 4 type since their removal is a common procedure to reduce overcrowding in the mouth. The age distribution of donated teeth is given in James *et al.* (2004, figure 2). For permanent teeth, the most common ages of extraction were between 11 and 14 years.

2.2. *Sources of elevated exposure to natural α-activity*

2.2.1. *Comparison of rural and urban areas.* The association previously observed between ^{210}Po levels in deciduous and permanent teeth and proximity to major motorways in the UK (Henshaw *et al.* 1995) was presumed to arise from the presence of ^{210}Pb emitted from vehicle exhausts, especially from diesel vehicles. Therefore, traffic pollution associated with the urban environment generally may lead to higher levels of ^{210}Po in the teeth of juveniles from urban compared with rural areas. Accordingly, three residence types were identified from Ordnance Survey maps:

- Rural: where the address was in population centres up to the size of a village, taken as less than 2000 people.
- Small town: where the address lay within a small town less than 80 000 inhabitants.
- Suburban and urban: encompassing addresses within a population centre of more than 80 000 inhabitants.

Mean α-activity concentrations in teeth were then grouped for each category. Given the investigation described below into the influence of domestic radon exposure on ^{210}Po in teeth, the authors were aware that the county of Devon was potentially in a separate category. The mean radon concentration in houses in Devon is around 10 times higher than the average 20 Bq m^{-3} for the UK and in some parts of the county indoor concentrations of several thousand Bq m^{-3} have been recorded. At the same time, Devon is a dominantly rural area with relatively low traffic density.

James *et al.* (2004) showed an association between α-activity concentration in teeth and post-eruption age. This has been examined further here in relation to residence type.

2.2.2. *Influence of motorways with respect to wind direction.* A map of the UK motorway network is shown in figure 1. Statistically significant associations between α-activity in teeth and residence within 10 km of the main north–south arterial routes of the M5/M6 and M1/A1 were found for deciduous but not for permanent teeth (Henshaw *et al.* 1995). For permanent teeth, the association became significant only within 2 km of motorways. It is of interest to consider the airborne transport of ^{210}Pb from exhaust emissions. To do this, ^{210}Po levels in permanent teeth with distance from motorways were compared in both the up- and downwind directions of the prevailing westerly to south-westerly wind across the UK. This was possible for the M5/M6, the M4 and the M25. Insufficient permanent teeth were available from donors living near the M1 and A1.

2.2.3. *Influence of radon.* Since ^{210}Po occurs at the end of the ^{222}Rn radioactive decay chain, domestic radon exposure should be considered as another potential source of increased ^{210}Po in teeth. Clemente *et al.* (1982, 1984) found an association between ^{210}Po in teeth and high cumulative radon exposure in persons living near to the Badgastein spa in Austria. It is of interest to determine whether such an association can be seen at much lower radon concentrations in UK homes.

Analysis of the variation in radon concentration by postcode in England carried out by the National Radiological Protection Board (Lomas *et al.* 1996) reveals a significant negative correlation between radon concentration and urban residence type ($r = -0.26$, $p < 0.0005$, $n = 251$). This indicates that the higher the level of urbanization of a region, the lower the mean domestic radon level. This effect is likely to be coincidental, with urban population sites centred in low radon regions purely by chance. However, it appears to confirm that an urban effect on α-activity concentration in teeth may directly conflict with any radon effect present. Therefore, in the present analysis, possible relationships between ^{210}Po activity concentration in teeth and radon concentration have been examined both for all individuals and for individuals by residence type. The analysis has been confined to England to match the data available in Lomas *et al.* (1996).

Figure 1. Principal motorways in the UK.

2.3. *Tooth analysis*

The analysis procedures were described in James *et al.* (2004). The outer enamel surface of permanent teeth was analysed by α-particle autoradiography using TASTRAK plastic track detectors, stored against each tooth for periods between 380 and 1120 days. James *et al.* determined that the activity on the outer enamel tooth surface was almost entirely composed of ^{210}Po.

The analysis used here was based on permanent teeth from 278 juveniles in 48 counties. As part of the geographical survey, over 6000 permanent teeth were collected. However, the number of teeth available for the present analysis was limited as 4000 teeth were used in radiochemical analyses to measure levels of plutonium (in batches of 50) and strontium (in batches of eight) across the UK (O'Donnell *et al.* 1997). For the remaining available teeth, their value was limited by inadequate information concerning the donor. For example, it was desirable to have the donor's date of birth rather than simply the age in years, and it was essential to have the full address, including the six- or seven-digit postcode. In selecting teeth near motorways, those donated were dominated by certain counties and regions, notably Devon, the West Midlands and Merseyside. There are counties from where only one tooth was obtained. Note that in most cases two teeth per donor were obtained because orthodontic premolar 4s are usually extracted in pairs. In some cases, four teeth were obtained from a donor. In all the measurements presented, where multiple teeth were analysed from a donor, an average activity concentration for the donor was obtained.

Alpha-particle autoradiographs of the outer surface of teeth were analysed by automated image analysis as described in Henshaw *et al.* (1994) and James *et al.* (2004). Repeat analyses using the Bristol image analysis system are generally well reproducible and grey-scale calibration slides are used to ensure consistency in the optical set-up. However, to minimize the possibility of systematic errors, all autoradiographs were analysed in one batch with careful checking of the optical set-up throughout.

3. Results

3.1. *Activity concentrations in teeth from urban, small town and rural areas*

Table 1 and figure 2a show the geometric mean activity concentrations in teeth from rural, small town and urban areas. The geometric mean was used throughout, since the distribution of activity concentration in teeth followed a log-normal distribution (James *et al.* 2004). When all counties were included, the mean activity concentration in urban areas was only marginally higher compared with rural areas. However, when considered alone, the activity concentration in 30 teeth obtained from Devon was unexpectedly high, given that it is mainly a rural area.

Table 1. Geometric mean ^{210}Po α-activity concentration on outer tooth surface enamel by residence type.

	Total α-activity concentration ($Bq\,kg^{-1}$)	
Residence type	Including Devon	Excluding Devon
Rural	7.83 +0.47/−0.45[a] 35	7.20 +0.49/−0.46 25
Small town	7.94 +0.38/−0.36 72	7.76 +0.37/−0.35 55
Urban	8.48 +0.25/−0.24 169	8.41 +0.25/−0.24 166

[a]Errors refer to the standard error in the ln distribution.

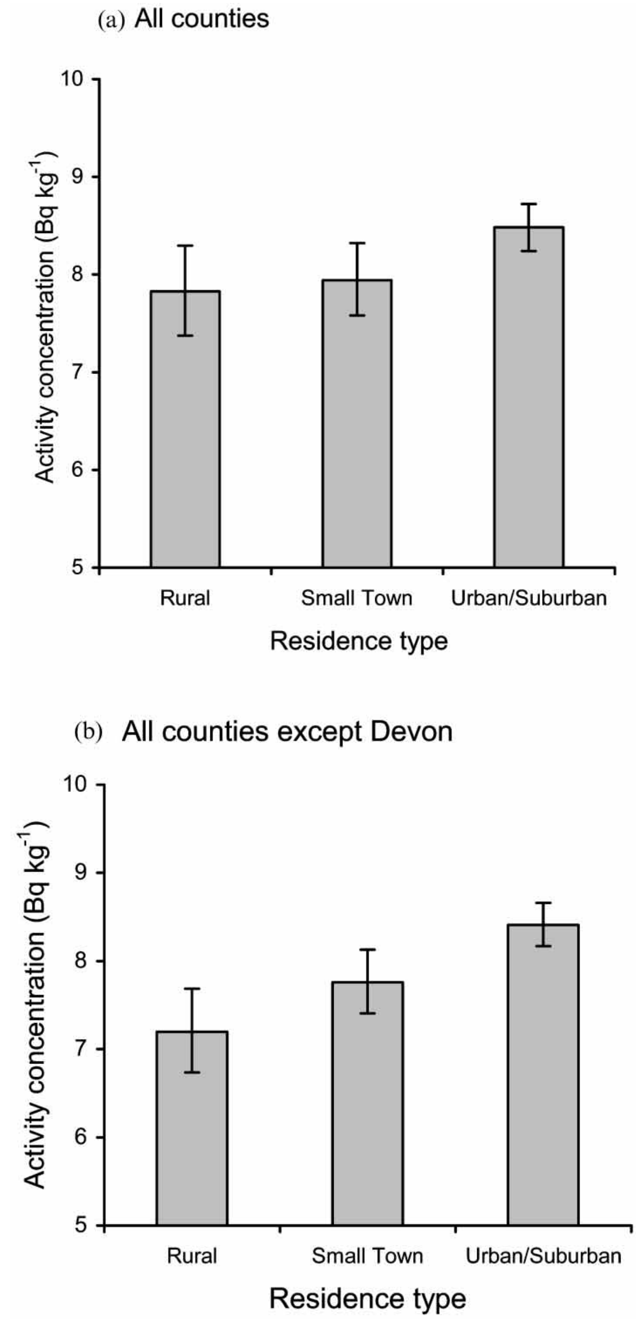

Figure 2. Geometric mean ^{210}Po α-activity concentrations in teeth by residence type: (a) all counties and (b) all counties, except Devon.

Table 2. Summary of correlation analysis between ^{210}Po α-activity concentration for permanent teeth, sorted by residence type and post-eruption tooth age.

Residence type	Pearson correlation coefficient, *r* (sample size)	*p*
Rural	0.01 (32)	0.94
Small town	0.02 (66)	0.90
Urban	0.26 (155)	0.003

As stated above, houses in Devon are known to have high mean indoor radon concentrations, which could confound the measurements with respect to urbanity. When this county was removed from the analysis, the difference in mean activity concentration between rural, small town and urban areas was more marked (figure 2b). The difference in mean activity concentration between rural and urban areas then become statistically significant ($p = 0.04$).

The association with post-eruption tooth age for the three residence types is shown in table 2. The association was significant only for urban areas ($p = 0.003$, $n = 155$). Note that in all of the results presented, *p* values were adjusted for multiple testing of variables as appropriate.

3.2. *Motorways*

Overall, there was no significant association between α-activity on the outer enamel surface of permanent teeth and proximity to the M5/M6, M4 or M25, either collectively or individually, confirming the earlier findings. However, when up- and down-wind distances were considered separately, there was a statistically significant association with respect to donors living downwind (on the easterly side) of the motorways (figures 3a and b, and table 3). To examine further the influence of traffic density, sections of the M5 and M6 that traverse urban areas were isolated from those traversing rural and small town areas. In the latter case, no significant association with downwind distance was observed, but a significant association was found for urban areas (figures 3c and d, and table 4).

3.3. *Radon*

Table 5 shows the correlation between α-activity concentration in teeth and domestic radon levels by residence type. For all residence types together, the association with radon was just short of statistical

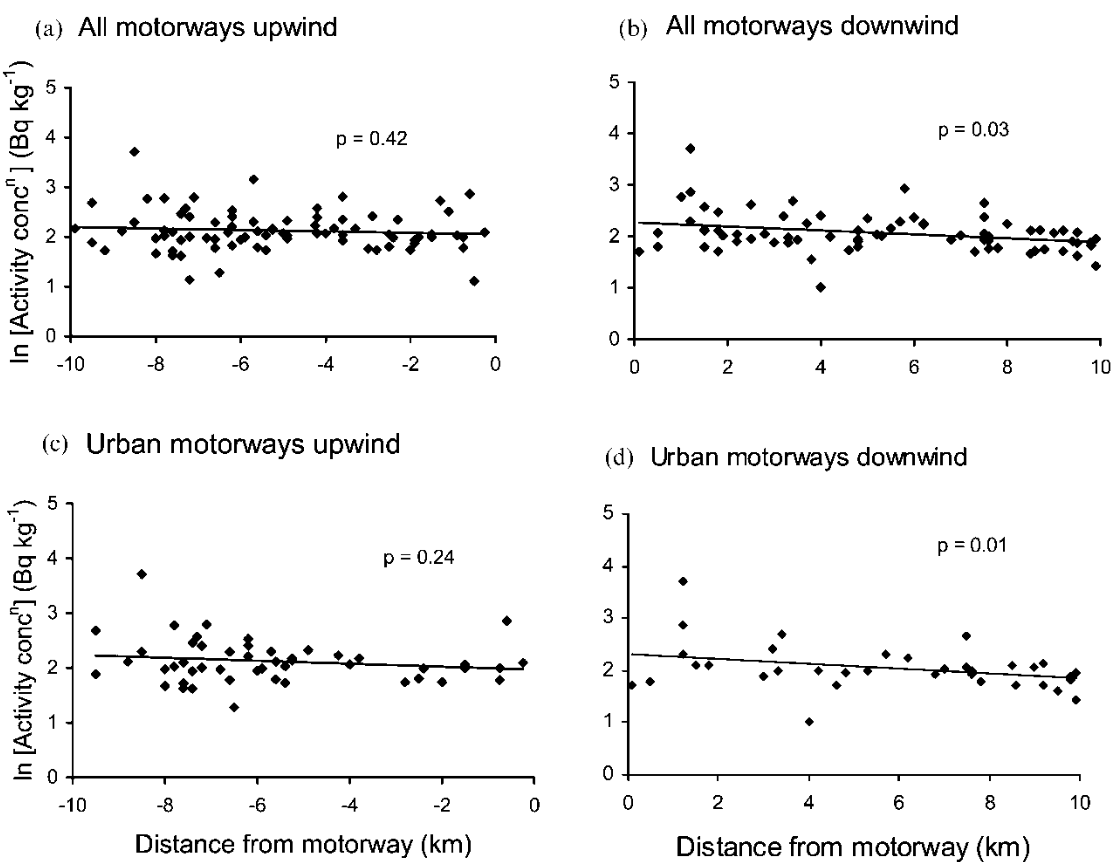

Figure 3. ^{210}Po α-activity in teeth with distance from motorways, upwind and downwind with respect to the prevailing west to south-westerly wind across the UK.

Table 3. Summary of correlation analysis between ln (^{210}Po α-activity concentration) and prevailing wind-related distance from residence to motorways.

Distance from motorway (km)	Pearson correlation coefficient, r (sample size)	p
Downwind		
<20	0.10 (116)	0.26
<10	−0.30 (73)	0.03
<5	−0.31 (36)	0.07
Upwind		
<20	0.03 (101)	0.80
<10	0.09 (80)	0.42
<5	0.18 (33)	0.31

Table 4. Summary of correlation analysis between ln (^{210}Po α-activity concentration) and distance from residence to motorways in urban areas only.

Distance from motorway (km)	Pearson correlation coefficient, r (sample size)	p
Urban residence type, downwind		
<20	0.07 (87)	0.53
<10	−0.40 (50)	0.01
<5	−0.32 (27)	0.11
Urban residence type, upwind		
<20	0.14 (59)	0.29
<10	0.17 (49)	0.24
<5	0.04 (14)	0.89

significance ($p = 0.08$). However, the corresponding associations for rural and small town residences were significant ($p = 0.04$ and 0.02, respectively). No association was found for urban residences. Figure 4 shows a plot of the data for non-urban residences.

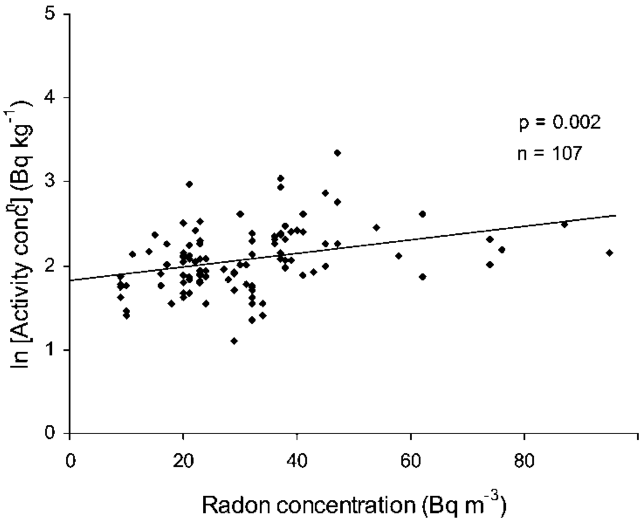

Figure 4. ^{210}Po α-activity in teeth versus mean indoor radon concentration by postcode. Each point represents an individual child.

4. Discussion

4.1. *Urban versus suburban and rural areas*

The higher levels of ^{210}Po in teeth from urban compared with rural areas are likely to be attributed to trace levels of ^{210}Pb emitted in vehicle exhausts, especially diesel exhausts. This is despite the elimination in recent years of the use of leaded petrol. Indeed, the observations closely follow those for stable lead in teeth where a substantial literature exists. For example, a number of authors have reported higher levels of stable lead in teeth from urban compared with suburban areas and suburban compared with rural areas (Grobler *et al.* 1984, 1985a, b, Brockhaus *et al.* 1988, Srivastava *et al.* 1992, Karakaya *et al.* 1996). The differences have variously been ascribed to traffic density, the proximity of major roads, and the degree of industrialization between urban, suburban and rural areas.

The association of α-activity in teeth with post-eruption age in urban areas provides a further indicator of higher uptake of ^{210}Po in urban areas. The result is similar to the findings of Lappalainen and Knuuttila (1981) and Steenhout and Pourtois (1981), who showed that uptake of stable lead in teeth, with respect to both the age of the individual and to post-eruption tooth age, occurs more rapidly in urban areas compared with rural areas.

Table 5. Summary of correlation analysis between ln (^{210}Po α-activity concentration) for permanent teeth, arranged by residence type and domestic radon concentration by postcode area/district.

Residence type	Pearson correlation coefficient, r (sample size)	p
All	0.15 (276)	0.08
Rural	0.41 (35)	0.04
Small town	0.32 (72)	0.02
Urban	0.03 (169)	0.65

4.2. *Upwind and downwind of motorways*

In Henshaw *et al.* (1995), statistically significant associations between α-activity in teeth and residence within 10 km of the M5/M6 and M1/A1 were found for deciduous but not for permanent teeth. For permanent teeth, the association became significant only when distances within 2 km were considered. Thus, the lack of an overall association with proximity to the M5/M6, the M4 and M25 agrees with the previous findings.

For the upwind/downwind analysis, statistical power was limited. Nevertheless, in the pooled data, an association was found with α-activity concentration in teeth downwind of motorways with respect to the prevailing west or south-westerly wind across the UK. This association is consistent with the airborne transport of vehicle exhaust pollution. It is interesting to note that in both Henshaw *et al.* (1995) and the present work, the significant associations extend to 10 km from the motorway. This is consistent with the view that a significant proportion of vehicle exhaust pollution exists in the form of ultrafine aerosols, around 200 nm in size, which can be carried comparatively large distances from their source and constitute a particular health hazard when inhaled, due to their ability to penetrate deeply into the lung (Seaton *et al.* 1995).

4.3. *Radon*

Toth *et al.* (1982) compared ^{210}Pb and ^{210}Po levels in teeth from those living in the country, in towns and in coal miners in Hungary. Higher levels were seen in the teeth from coal miners and this was related to cumulative radon exposure in coal mines. Clemente *et al.* (1982, 1984) found an association between ^{210}Po in teeth and high cumulative radon exposure in persons living near to the Badgastein spa in Austria. In the centre of the town, water from thermal springs originates with a mean radon concentration of 1.5×10^6 Bq m^{-3}. The rate of uptake of ^{210}Po was 1.2 mBq g^{-1} per working level month (WLM) lifetime exposure.

The association with domestic radon concentration at the much lower levels in the UK is unexpected. The magnitude is approximately equal to that from traffic density at the average UK radon exposure of 20 Bq m^{-1}, but becomes more important at higher levels of indoor radon. This agrees with a similar observation by Anttila (1987) who measured stable ^{206}Pb, the end-point of the ^{222}Rn decay chain, on the enamel surface of Finnish children living in high and low radon areas. By coincidence, the high radon area was a rural region while the low radon area was an urban industrialized region. ^{206}Pb levels were higher in teeth from the high radon area and appeared to predominate over the difference between urbanization between the two regions.

4.4. *Relative importance of oral, inhalation and ingestion uptake with respect to ^{210}Pb in outer tooth enamel*

The question arises as to the dominant method of uptake of ^{210}Pb in outer tooth enamel. Airborne concentrations of ^{210}Pb are very low, of the order 1–2 mBq m^{-3} of air (Hötzl and Winkler 1987). On inhalation, most of the radon short-lived decay products deposit in the tracheobronchial region of the lung, the site where most decay to ^{210}Pb. The latter may enter the bloodstream directly or may be cleared by the mucociliary escalator, entering the bloodstream by gut transfer. This would therefore suggest that the excess ^{210}Pb in teeth associated with domestic radon exposure is absorbed into the tooth surface primarily by inhalation rather than orally. Henshaw *et al.* (1994) noted that on the outer tooth surface, the activity was higher on the cement below the gum line compared with the enamel above the gum line. Together, this suggests direct absorption from blood below the gum line and absorption from saliva above the gum line. Such systemic uptake would also apply to the enhanced activity at bone surfaces reported in Salmon *et al.* (1994, 1995).

On the other hand, the association with traffic pollution suggests direct exposure to airborne ^{210}Pb from its presence in oils, so in this case both the oral and inhalation routes of uptake could apply.

The results also indicate that together both routes are more important than ingestion as a source of ^{210}Pb on the outer enamel of teeth. For the skeleton, Salmon *et al.* (1998) modelled the relative contribution of inhalation and ingestion uptake of ^{210}Pb. The authors concluded that in the absence of radon, inhalation accounts for only 5% of uptake in the UK compared with 50% for Illinois, USA. The average UK indoor radon concentration of 20 Bq m^{-3} was estimated to add an additional 1.8% only to the total skeletal burden compared with 3.9% for Illinois. The findings here in teeth testify to the importance of oral and/or inhalation uptake since dietary sources of ^{210}Pb would be expected to be approximately uniform across the UK. Thus, in table 1, ^{210}Po levels in teeth are 17% higher in urban compared with rural areas, and according to figure 4, mean levels of ^{210}Po in teeth at zero, 20 and 100 Bq m^{-3} are, respectively, 6.2, 7.2 and 13.6 Bq kg^{-1}.

4.5. *Implications for dose assessment*

Manea-Krichton *et al.* (1991) showed a correlation between stable lead in permanent teeth and that in the rib and ulna of adults. Similarly, Lovaas and Hursh (1968) found that in adults ^{226}Ra and ^{210}Pb levels in permanent teeth correlated well with those in various bones. Salmon *et al.* (1994, 1995) found enhanced levels of ^{210}Po at endosteal bone surfaces similar to that seen in teeth. In the present work, the findings of associations between ^{210}Po in outer enamel of permanent teeth from juveniles and certain sources of enhanced exposure may help refine estimates of levels in bone and the subsequent high-LET dose to bone marrow.

The National Radiological Protection Board (NRPB) has estimated that of the total high-LET equivalent dose to bone marrow in children, α-emission from ^{210}Po contributes around 180 μSv y^{-1} in the UK (NRPB-R276 1995, table 5.6). The Committee on Medical Aspects of Radiation in the Environment has suggested that 34% of the incidence of childhood leukaemia is linked to natural background radiation, 20% from low-LET β- and γ-radiation, and 14% from high-LET α-radiation (COMARE 1996). Partitioning the dose to red bone marrow between the principal emitters using the NRPB dose estimates suggests that 5% of childhood leukaemia may be linked to ^{222}Rn, ^{220}Rn and their short-lived α-emitting decay products, and 9% to longer-lived emitters, especially ^{210}Po. In the present work, it is the concentration of ^{210}Po specifically on the outer surface of tooth enamel, as opposed to the tooth volume, that appears to reflect integrated exposure (James *et al.* 2004). Further effort is now needed to determine whether ^{210}Po in outer tooth enamel constitutes a useful indicator of integrated exposure in bone and in turn the high-LET dose to bone marrow.

With regard to ^{210}Po as a component of vehicle pollution, it should be emphasized that vehicle exhausts contain known chemical carcinogens especially benzene and the polycyclic-aromatic hydrocarbons such as benzo[α]pyrene (Nisbet and LaGoy 1992, Glass *et al.* 2003). Indeed, some studies have specifically reported an association between traffic density and childhood leukaemia risk (Savitz and Feingold 1989, Knox and Gilman 1997, Nordlinder and Järvholm 1997, Feychting *et al.* 1998, Harrison *et al.* 1999, Pearson *et al.* 2000).

4.6. *Form of the relationship between ^{210}Po in teeth, distance from motorways and radon concentration*

In figures 3 and 4, it is evident that the scatter in the data is such that proximity to motorways and domestic radon concentration are poor indicators of ^{210}Po activity concentration in individual teeth. However, for purposes of radiation protection and risk analysis, the data may be used to predict average levels in teeth for young populations exposed to these sources.

Thus, for motorways in general, the relationship between downwind distance and ^{210}Po in teeth may be given by:

$$\begin{aligned}&\ln(\text{total }\alpha-\text{activity concentration}) = \\ &(-0.038\times\text{motorway distance})+2.26\end{aligned} \tag{1}$$

with 95% confidence limits in the slope between −0.068 and −0.009.

For urban sections of motorway the corresponding relationship is:

$$\begin{aligned}&\ln(\text{total }\alpha-\text{activity concentration}) = \\ &(-0.053\times\text{motorway distance})+2.35\end{aligned} \tag{2}$$

with 95% confidence limits in the slope between −0.088 and −0.018.

For domestic radon exposure the relationship may be given by:

$$\begin{aligned}&\ln(\text{total }\alpha-\text{activity concentration}) = \\ &(0.008\times\text{domestic radon concentration})+1.81\end{aligned} \tag{3}$$

with 95% confidence limits in the slope between 0.004 and 0.012.

In equations (1–3), distance is in kilometres, activity concentration in Bq kg^{-1} and radon concentration in Bq m^{-3}. Note again that the effects of domestic radon exposure on ^{210}Po levels in teeth on average are similar to those for traffic density.

Finally, it has not been possible to carry out a bivariate analysis of exposure to urban pollution and radon together. This would require modelling of urbanity as a continuous variable, which would be difficult in the present data set.

5. Conclusions

In the UK, geographical variations have been found in the level of ^{210}Pb-supported ^{210}Po on the outer enamel surface of permanent teeth extracted from juveniles for orthodontic purposes. These variations relate to exposure to urban pollution, living downwind of motorways and domestic radon exposure. The relationship with downwind distance from motorways is based on limited statistics. The magnitude of the association with radon is approximately equal to that from traffic density at the average UK radon exposure of 20 Bq m^{-1}, but becomes more important at higher levels of indoor radon.

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teeth donated from children and juveniles from the UK, the Republic of Ireland and Continental Europe.

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