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An unexpected rise in strontium-90 in US deciduous teeth in the 1990s

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Abstract

For several decades, the United States has been without an ongoing program measuring levels of fission products in the body. Strontium-90 (Sr-90) concentrations in 2089 deciduous (baby) teeth, mostly from persons living near nuclear power reactors, reveal that average levels rose 48.5% for persons born in the late 1990s compared to those born in the late 1980s. This trend represents the first sustained increase since the early 1960s, before atmospheric weapons tests were banned. The trend was consistent for each of the five states for which at least 130 teeth are available. The highest averages were found in southeastern Pennsylvania, and the lowest in California (San Francisco and Sacramento), neither of which is near an operating nuclear reactor. In each state studied, the average Sr-90 concentration is highest in counties situated closest to nuclear reactors. It is likely that, 40 years after large-scale atmospheric atomic bomb tests ended, much of the current in-body radioactivity represents nuclear reactor emissions.

Keywords: Radiation; Strontium-90; Nuclear reactors; Deciduous teeth (baby teeth)

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1. Introduction

Since man-made fission products were first released into the environment in the mid-1940s, determining in vivo levels of these radioisotopes has challenged scientists. Hundreds of radioisotopes are created in nuclear weapon detonations and in nuclear reactor emissions. Many of these are short-lived, and therefore highly unlikely to track in vivo. Collecting samples of longer-lived isotopes often involves invasive processes such as autopsies and biopsies, making collection of significant samples time-consuming and costly.

In the US, whose government conducted 206 atmospheric tests of nuclear weapons from 1946 to 1962 (100 in Nevada, 106 in the South Pacific) (Norris and Cochran, 1994), the federal government instituted programs measuring strontium-90 (Sr-90) concentrations in vertebrae. One focused on deceased adults (begun 1954, 3 cities, ~50 bones per year) (Klusek, 1984), while the other included deceased children and adolescents (begun 1962, 30 cities, ~300 bones per year) (Baratta et al., 1970). Both showed increases to a peak in 1964, just after the Partial Test Ban Treaty was signed, and a dramatic decline in the mid- and late 1960s.

The largest-scale US program studying in-body radioactivity was conducted in St. Louis. Kalckar suggested that large numbers of deciduous teeth could be collected and tested to examine the buildup of fallout from bomb tests (Kalckar, 1958). A coalition of St. Louis medical/scientific professionals and citizens collected over 300 000 teeth from local children from 1958 to 1970. Results from St. Louis were similar to the two bone programs, i.e.

- A 55-fold rise in average millibecquerels (mBq) of Sr-90 per gram calcium at birth (7.4–408.1) took place for 1949–1950 births (before large-scale tests began) to 1964 births (just after the largest-scale bomb tests ended).
- A 50% decline in Sr-90 concentrations in St. Louis fetal mandibles occurred from 1964 to 1969 births. This far exceeded the expected 9% reduction suggested by the 28.7 year half-life of Sr-90 (Rosenthal, 1969).

After the bone and tooth studies showed such a rapid post-1964 decline, federal funding was terminated for each program, and work ceased. The tooth study ended in 1970, the child bone study in 1971 and the adult bone study in 1982.

The US studies were accompanied by similar international efforts. Each independently confirmed the American findings of rapid increases in teeth until 1964, including studies in Czechoslovakia, Denmark, Finland and Scotland (Santholzer and Knaifl, 1966; Aarkrog, 1968; Rytomaa, 1972; Fracassini, 2002). Another study in Finland duplicated the rapid post-1964 plunge in Sr-90 (Kohlehmainen and Rytomaa, 1975). No nation maintained an ongoing program, but after the Chernobyl accident, reports from Germany, the Ukraine and Greece documented a substantial rise in Sr-90 in baby teeth after the April 1986 disaster (Scholz, 1996; Kulev et al., 1994; Stamoulis et al., 1999). Another study examined Sr-90 in teeth from children who lived proximate to the Sellafield nuclear installation in northwestern England; results are addressed in Section 4 (O'Donnell et al., 1997).

With no program of in vivo radioactivity to gauge the burden on the body, levels in the environment can be used as a proxy measure. In the past, patterns of Sr-90 in baby teeth were roughly equivalent to those of Sr-90 in milk (Rosenthal et al., 1964). The US government (beginning 1957) began publicly reporting monthly levels of a variety of radionuclides in milk and water in 40–60 US locations. However, a number of these radioisotopes, including Sr-90, strontium-89, cesium-137, barium-140 and iodine-131 were discontinued in the early 1990s (National Air and Radiation Environmental Laboratory, 1975–2001).

One measure that is still publicly reported is the concentration of gross beta particles in precipitation. A reduction in average beta levels reversed after 1986–1989. While the most recent 4-year period still features incomplete data, thus far the increase from 1986–1989 to 1998–2001 has been 53.8%. This difference is significant at $P<0.0001$,

Table 1
Trend in gross beta in precipitation in average millibecquerels per liter of water in 60 US cities, 1978–2001

4-year period	Months available	Number of measurements	Average beta[a]	Percent change, 1986–1989 to 1998–2001[b]
1978–1981	36	640	211	
1982–1985	48	1299	63	
1986–1989	46[c]	1845	58	
1990–1993	48	1892	59	
1994–1997	48	1696	63	
1998–2001	27	836	89	+53.8% ($P<0.0001$)

The *P* value indicates that the chance that the increase is due to random chance is fewer than 1 in 10 000. *Source:* Environmental Protection Agency, Environmental Radiation Data, quarterly volumes.

[a] Average millibecquerels of gross beta per liter of precipitation (reported by EPA as picocuries; to convert to millibecquerels, multiply by 37). Before 1996, figures were reported as nanocuries per meter squared at a particular depth (in millimeters); to convert to pCi/l, multiply nCi per meter squared times 1000, then divide by millimeters; then multiply by 37 to obtain millibecquerels.

[b] Calculation of change beginning with lowest average (1986–1989) to most current.

[c] Excludes May and June 1986, heavily affected by short-lived Chernobyl fallout.

i.e. the probability of this increase due to random chance is less than 1 in 10 000 (Table 1).

The lack of an ongoing program measuring in vivo radioactivity levels and an unexpected, sustained rise in environmental beta concentrations warrant a resumption of testing in vivo Sr-90 and perhaps other radioisotopes, first instituted during the era of atmospheric nuclear weapons testing.

In 1996, the Radiation and Public Health Project (RPHP) began a study of Sr-90 levels in deciduous teeth, focused on persons living near nuclear reactors. The goal of this project was to build a current database of in vivo radioactivity documenting Sr-90 patterns and trends. While Sr-90 is just one of hundreds of radioisotopes from fission, it can be used as a proxy for all fission products, especially those with extended half-lives.

2. Materials and methods

Earlier reports addressed methods used and initial findings from the baby tooth study (Gould et al., 2000a,b; Mangano et al., 2000). These teeth were processed using a scintillation counter from the University of Waterloo in Ontario, Canada. In June of 2000, RPHP leased a Perkin-Elmer 1220-003 Quantulus Ultra Low-Level Liquid Scintillation Spectrometer. Introduced in 1995, only approximately 15–20 units are now in use in the US (Laxton, Mark, Perkin-Elmer Life Sciences Inc., 549 Albany Street, Boston MA 02118. Personal correspondence, May 9, 2002).

The new counter is located on the premises of REMS, Inc., a radiochemistry laboratory in Waterloo, and not at the University of Waterloo, thus changing the level of background radiation. Also, the method of removing organic material from the teeth was changed by treating them with hydrogen peroxide prior to grinding them into powder. This procedure proved to be more effective in allowing light produced in the liquid scintillation fluid by the beta particles emitted by the Sr-90 and its daughter product, Yttrium-90, to reach the photomultipliers. This greater efficiency is caused partly by shifting the spectrum of the light emitted by the scintillation fluid. As a result of these changes (the counter, its location, level of background radiation and method of cleaning teeth), the efficiency of detecting the very low radioactivity in single teeth was more than doubled overall. However, the data lack a consistent factor that could be used to analyze teeth from both counters together. Thus, this report will be based solely on the 2089 deciduous teeth tested after June 2000.

RPHP sends teeth to REMS for testing, and Sr-90 levels are measured individually. Lab personnel are blinded about all information concerning each tooth, that is, they know nothing about character-

Table 2
Average millibecquerels of Sr-90 per gram calcium (at birth) in deciduous teeth from St. Louis, 1954 and 1959 births (test for internal consistency)

Batch		Average Sr-90[a]	% 1959 over 1954	Counting error	95% confidence interval
#1	1954	61		±10	41–81
	1959	121	+98	±13	95–147
#2	1954	65		±11	43–87
	1959	124	+90	±14	96–152

[a] Average millibecquerels of Sr-90 per gram of calcium.

istics of the tooth donor. This blinding helps assure objectivity in results. The laboratory measures the concentration of Sr-90 by calculating the current activity (in mBq) of Sr-90 per gram of calcium in each tooth (mBq Sr-90/g Ca). (See Appendix A for more specific technical procedures.) The strontium-to-calcium ratio has been used in the St. Louis study in the 1960s, and all other recent baby tooth studies mentioned earlier.

The laboratory returns results to RPHP staff, who converts the ratio to that at birth, using the Sr-90 half-life of 28.7 years. The Sr-90/Ca ratio for a single tooth is not a precise number because a typical baby tooth is small in mass. The counting error for each tooth is plus or minus 26 mBq, and somewhat less for the larger teeth.

RPHP conducted several tests to assure the inter-laboratory reliability and internal consistency of its results. It selected 10 teeth from persons born in 1954 in St. Louis that were tested both by REMS and the University of Georgia Center for Applied Isotope Studies, which operates three counters of the same model. REMS dried the 10 teeth and ground them into a powder. After testing for Sr-90 levels, the entire batch was sent to the University of Georgia, which tested a dissolved solution of teeth. Both labs were blinded from each other's results. The data were relatively comparable. REMS' average was 65 mBq Sr-90/g Ca (CI=43–87), while University of Georgia's tally was 79 mBq/g Ca (CI=56–102).

REMS also performed a second test, for internal consistency. Prior results from the St. Louis study indicated that average 1959 Sr-90 levels were considerably higher than those for 1954, due to buildup in bomb test fallout. RPHP split two samples of 10 teeth, each into two batches, and asked REMS to calculate average Sr-90 levels separately. Results, shown in Table 2, documented the 1959 average to be 98 and 90% higher than the 1954 average. Confidence intervals showed considerable overlap, indicating that study results are consistent both internally and with the earlier St. Louis study.

A third test for accuracy involved several dozen teeth from persons born in the Philippines Islands 1991–1992. This area has never had a nuclear reactor (for weapons, power or research). It may have received fallout from Chinese atmospheric bomb tests, but there were many fewer of these than US tests. Chinese atmospheric tests ended in 1980, and the last below-ground test occurred in 1993. Thus, Philippino teeth should contain lower concentrations of this radioisotope than American teeth.

Thirteen teeth of Philippino children born in 1991 and 1992 were tested. The average concentration at birth was 75 mBq Sr-90/g Ca, or 41% lower than the 127 mean level for American children born in those years.

RPHP collects teeth through voluntary donations, mostly from parents of children who have recently shed a deciduous tooth. Donors submit teeth in envelopes containing identifying information on the child and parents. RPHP staff assigns each tooth a unique tracking number. The group sent nearly 100 000 unsolicited letters appealing for tooth donation to families with children age 6–17. These mailings occurred in California (Sacramento and San Luis Obispo counties), Florida (Dade and Port St. Lucie counties) and New York (Rockland and Westchester counties). Families

Table 3
Average millibecquerels of Sr-90 per gram calcium in deciduous teeth (at birth) by state (all persons and persons born after 1979)

State	Teeth	Average Sr-90[a]	Counting error
All persons			
PA	133	155	±14
Oth	492	146	±7
NY	557	141	±6
NJ	271	139	±9
FL	485	131	±6
CA	151	114	±10
TOT	2089	139	±3
Persons born after 1979			
PA	130	154	±14
NY	534	138	±6
FL	471	130	±6
Oth	417	130	±6
NJ	244	125	±8
CA	138	108	±10
TOT	1934	132	±3

See Appendix B for explanation of error calculation.
[a] Average millibecquerels of Sr-90 per gram of calcium.

receiving letters were randomly selected by zip code in each county, that is, every *n*th family in each zip code received a letter. Just over 1% of these mailings were returned with a baby tooth enclosed.

Teeth are geographically classified by the zip code where the mother resided during pregnancy, rather than the current residence. The large majority of Sr-90 uptake in a baby tooth occurs during the fetal and early infant periods (Rosenthal, 1969), making zip code during pregnancy the appropriate geographic identifier.

Other teeth were collected from persons who became familiar with the project through media articles and stories, and through the group's web site. Thus, the teeth are not necessarily representative of the US population at large. The vast majority is concentrated in only five states (California, Florida, New Jersey, New York and Pennsylvania), near nuclear reactors. Most were donated from children who have just recently lost a tooth, or those between age 5 and 13. Despite these shortcomings, the large number of teeth will enable meaningful analysis of average Sr-90 concentrations to be performed; and any major variations—by birth year, by state, etc.—will likely be discernible.

3. Results

3.1. By state

A total of 2089 teeth were tested for Sr-90, and are discussed in this paper (another 1335 had been tested previously using a different scintillation counter and method). As discussed, the two sets of results are each internally consistent, but not comparable with each other because of differences in the counter, its location, level of background radiation and method of cleaning teeth, so only the last 2089 teeth are used. Of these, 1592 (77%) were from children born in the five states mentioned earlier, each with at least 133 teeth studied. No other state has more than 34 teeth. Table 3 shows the comparative average Sr-90 concentrations by state.

Table 3 also displays averages by state only for persons born after 1979. The large buildup from above-ground nuclear weapons tests reached a peak in 1964, and fell by approximately half over

Table 4
Average millibecquerels of Sr-90 per gram calcium in deciduous teeth (at birth) by proximity to nuclear power plants (persons born after 1979)

Nuclear power plant, location	Proximate counties	Average Sr-90[a] (No. teeth)		% Difference average Sr-90
		Proximate	Other state	
Indian Point, Buchanan NY (2 reactors, startup 1973, 1976)	Putnam, Rockland, Westchester, NY	164 (217) ±11	121 (317) ±7	+35.8% $P<0.001$
Limerick, Pottstown PA (2 reactors, startup 1984, 1989)	Berks, Chester, Montgomery, PA	168 (98)[b] ±17	110 (32) ±20	+53.2% $P<0.03$
Turkey Point, Florida City FL (2 reactors, startup 1972, 1973)	Broward, Dade, Palm Beach, FL	129 (350) ±7	93 (24) ±20	+38.6% $P<0.08$
St. Lucie, Hutchinson Island FL (2 reactors, startup 1976, 1983)	Indian River, Martin, St. Lucie, FL	143 (97) ±15	93 (24) ±20	+53.8% $P<0.04$
Oyster Creek, Forked River NJ (1 reactor, startup 1969)	Monmouth, Ocean, NJ	128 (169) ±10	119 (75) ±14	+8.1%
Diablo Canyon, Avila Beach CA (2 reactors, startup 1984, 1985)	San Luis Obispo, Santa Barbara, CA	127 (50)[b] ±19	97 (88) ±11	+30.8%

Counting error listed for each sample of teeth. See Appendix B for explanation of standard error calculation, Appendix C for significance testing. *Source:* US Nuclear Regulatory Commission (www.nrc.gov), obtained August 12, 1999, for reactor locations and startup dates.

[a] Average millibecquerels of Sr-90 per gram of calcium.

[b] In three counties near Limerick, 94 of 98 teeth were from persons born after startup (average 168). In two counties near Diablo Canyon, 47 of 50 teeth were from persons born after startup (average 135).

the next 5 years. Thus, continued decline of Sr-90 from bomb test fallout should have reached a level approaching zero by about 1980, and averages should largely represent current sources of this radionuclide. Average Sr-90 concentration for all teeth was 132 mBq Sr-90/g Ca, and state averages ranged from a high of 154 in Pennsylvania to a low of 108 in California.

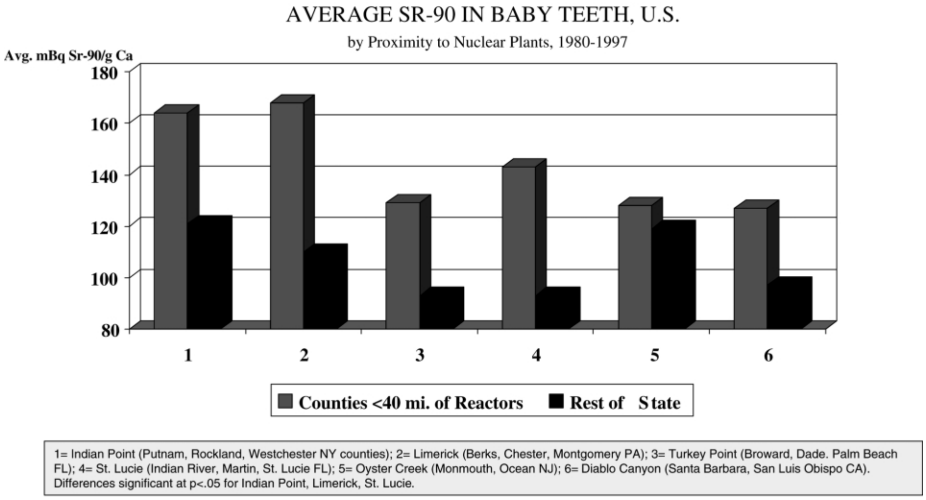

Fig. 1. Average Sr-90 in baby teeth, US, by proximity to nuclear plants (persons born 1980–1997).

Table 5
Average Sr-90 concentration (by birth year), US, in deciduous teeth (at birth)

Birth year	No. teeth	Average Sr-90[a]	Counting error
1954–1957	6	191	±78
1958–1961	8	331	±117
1962–1965	8	351	±124
1966–1969	17	272	±66
1970–1973	38	222	±36
1974–1977	38	211	±34
1978–1981	78	140	±16
1982–1985	172	140	±11
1986–1989	532	109	±5
1990–1993	836	132	±5
1994–1997	346	162	±9
% Change, 1986–1989 to 1994–1997			+48.5% $P<0.0001$

Note: Most teeth are from states of CA, FL, NJ, NY and PA. See Appendix B for explanation of error calculation, Appendix C for significance testing.
[a] Average millibecquerels of Sr-90 per gram of calcium.

3.2. By proximity to nuclear reactors

The question of whether those living closest to nuclear plants have higher burdens of radioactivity was addressed. Most teeth from residents close to nuclear plants—defined as counties situated mostly or completely within 40 miles—include six nuclear installations, described in Table 4 and Fig. 1. Average Sr-90 concentrations are compared with those from all counties in the remainder of the state, which are farther from reactors.

For each of the six areas, the local average of Sr-90 exceeded that for the remainder of the state. Three of the six differences are significant at $P<0.05$, with one other of borderline significance ($P<0.08$). Aside from a 8.1% excess near the Oyster Creek plant in central New Jersey, average Sr-90 concentrations near the other five reactors ranged from 30.8 to 53.8% above other counties in these states. Two parts of California can be considered relatively unexposed control areas. One is composed of Sacramento and El Dorado, close

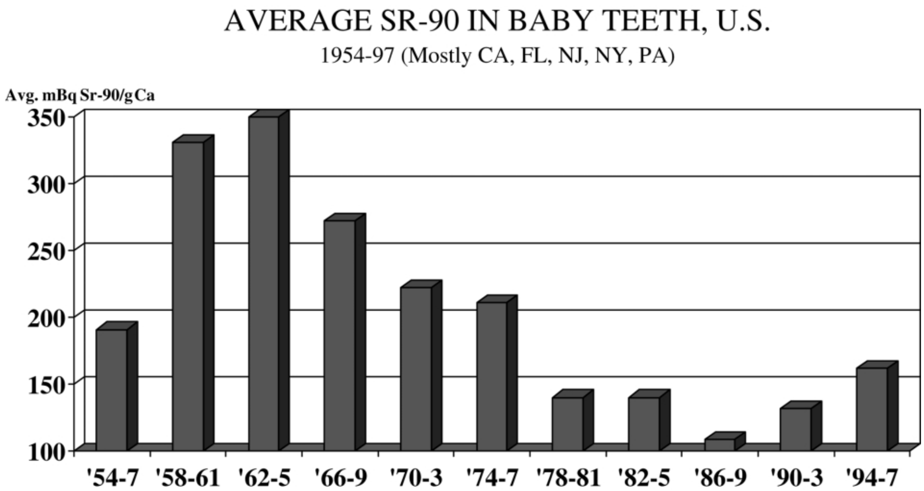

Fig. 2. Average Sr-90 in baby teeth, US, 1954–1997 (mostly CA, FL, NJ, NY, PA).

Table 6
Trend in Sr-90 concentration after 1981 in deciduous teeth, at birth by birth year, by state

Birth year	No. teeth	Average Sr-90[a]/counting error		No. teeth	Average Sr-90[a]/counting error	
California				*Florida*		
1982–1985	12	104	±31	63	133	±17
1986–1989	50	93	±14	102	112	±11
1990–1993	53	112	±16	192	127	± 9
1994–1997	20	139	±32	99	153	±16
% Change 1986–1989 to 1994–1997			+50.2%			+36.3% $P<0.04$
New Jersey				*New York*		
1982–1985	19	117	±27	41	153	±24
1986–1989	71	105	±14	142	120	±10
1990–1993	109	132	±13	237	128	± 9
1994–1997	39	144	±23	104	184	±18
% Change 1986–1989 to 1994–1997			+36.5%			+53.6% $P<0.002$
Pennsylvania				*All other*		
1982–1985	6	293	±120	31	134	±24
1986–1989	32	125	±23	135	100	±9
1990–1993	52	152	±21	193	141	±10
1994–1997	36	160	±27	48	159	±23
% Change 1986–1989 to 1994–1997			+27.7%			+59.0% $P<0.02$

See Appendix B for explanation of error calculation, Appendix C for significance testing.
[a] Average millibecquerels of Sr-90 per gram of calcium.

to the Rancho Seco nuclear plant, which closed in June 1989. The other is the San Francisco Bay area, which lies approximately 80 miles from Rancho Seco and 210 miles from the Diablo Canyon plant. The 50 teeth from persons born after 1979 near Diablo Canyon have the highest Sr-90 concentration in the state (127 mBq/g Ca), followed by those near the closed Rancho Seco plant (106 mBq/g Ca, 27 teeth), and the San Francisco Bay area (87 mBq/g Ca, 23 teeth).

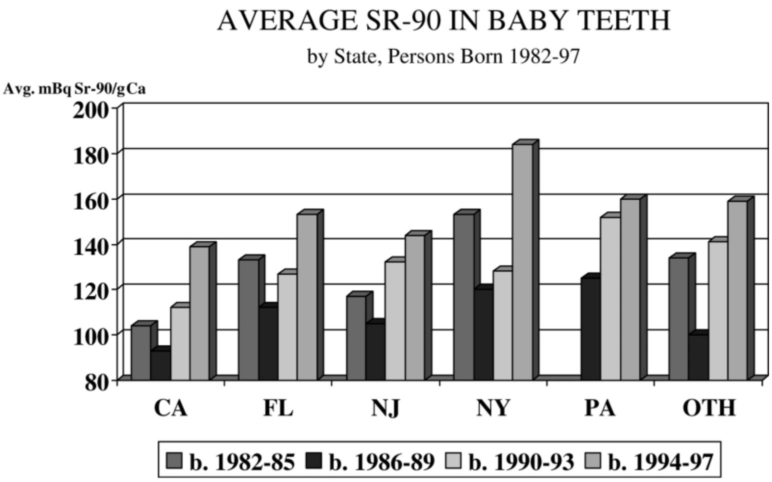

Fig. 3. Average Sr-90 in baby teeth, by state (persons born 1982–1997).

3.3. Temporal trends—total

Temporal trends in average in vivo Sr-90 concentrations were also analyzed. The earlier St. Louis study documented a 50% decline in average Sr-90 concentration in fetal mandibles in the 5 years after the Limited Test Ban Treaty went into effect (Rosenthal, 1969). The adult bone (vertebrae) program administered by the US government showed a similar decline, followed by a more modest reduction since the mid-1970s; this program was small in scope, and ceased in 1982 (Klusek, 1984). The teeth analyzed in this report represent persons born primarily in the 1980s and 1990s, providing data for a population not heretofore addressed.

Table 5 and Fig. 2 display the trend in average Sr-90 concentrations from the mid-1950s to the late 1990s. The trends established by earlier analyses (a rise until the mid-1960s followed by a decline until the early 1980s) were duplicated, even with a limited number of teeth studied prior to 1980. The new findings for those children born after 1981, who contributed 91% of all samples in the study, showed that the decline continued until the period 1986–1989. Four-year birth cohorts are used here to maximize numbers of teeth and smooth trends. In 1986–1989, the lowest average Sr-90 concentration in the study was observed (109 mBq Sr-90/g Ca), well below the 351 mBq Sr-90/g Ca observed in the mid-1960s. This long-term decline was followed by an increase of 48.5% in the next two 4-year periods, ending with an average of 162 mBq Sr-90/g Ca for the 1994–1997 birth cohort ($P<0.0001$). Although trends for individual years are less reliable due to fewer teeth, the lowest average was reached in 1986 (94 mBq Sr-90/g Ca for 76 teeth) and the highest average thereafter occurred in 1996 (195 mBq Sr-90/g Ca for 30 teeth), an increase of 107% ($P<0.007$). Only six teeth for births after 1996 have been analyzed to date.

3.4. Temporal trends—by state

The unexpected and abrupt reversal of declines in Sr-90 concentration in US baby teeth takes on greater meaning when data from each state are analyzed. 'National' data essentially include only five states, and thus may or may not be representative of the entire US. However, for post-1981 births, each of the five states duplicates the same trend; a reduction to a post-Test Ban low in 1986–1989, followed by two successive increases in the following 4-year periods. The geographic disparity of these areas suggests that the trend may apply nation-wide, at least in areas near nuclear reactors, from which most study teeth were donated. Table 6 and Fig. 3 display these consistent trends, which also occurred for the 'all other' categories (teeth from children in areas other than the five focus states). Rises during the 1990s vary from 27.7 to 59.0%. Increases in Florida, New York and 'other' states are significant ($P<0.05$).

3.5. Temporal trends—by counties

The trends in states were also consistent for the counties (or clusters of counties) that donated the most teeth to the study (Table 7). These include Monmouth/Ocean County, NJ (closest to the Oyster Creek plant), Dade County, FL (site of the Turkey Point plant) and Putnam/Rockland/Westchester Counties, NY (which converge at the Indian Point plant). Increases from 1986–1989 to 1994–1997 ranged from 49.8 to 55.7%, with the Florida and New York counties achieving statistical significance ($P<0.05$). The only slight exception to this trend was that all of Monmouth/Ocean County's increase took place in the early 1990s.

4. Discussion

The US has conducted no official program measuring in vivo levels of fission products for over 20 years. This report introduces current data on patterns and trends of Sr-90 concentration in US baby teeth, mostly near nuclear power installations. The average concentration of Sr-90 was 132 mBq Sr-90/g Ca for all children born after 1979, when in vivo Sr-90 remaining from atomic

Table 7
Trend in Sr-90 concentration after 1981 in deciduous teeth (at birth) by birth year, by county (counties with the largest sample sizes)

Birth year	No. teeth	Average Sr-90[a]	Counting error
Dade County FL			
1982–1985	47	141	±21
1986–1989	57	94	±13
1990–1993	106	124	±12
1994–1997	43	141	±22
% Change 1986–1989 to 1994–1997			+50.6% $P<0.057$
Monmouth, Ocean Counties NJ			
1982–1985	13	150	±40
1986–1989	44	93	±14
1990–1993	76	140	±16
1994–1997	31	139	±25
% Change 1986–1989 to 1994–1997			+49.8%
Putnam, Rockland, Westchester Counties NY			
1982–1985	17	202	±50
1986–1989	43	135	±21
1990–1993	101	148	±15
1994–1997	52	211	±30
% Change 1986–1989 to 1994–1997			+55.7% $P<0.04$

See Appendix B for explanation of error calculation, Appendix C for significance testing.
[a] Average millibecquerels of Sr-90 per gram of calcium.

bomb tests should approach 0.[5] This concentration is lower than that in those born before 1980, when bomb test fallout accounted for a substantial proportion of in vivo radioactivity. However, it exceeds the levels before the large-scale testing began in 1951 in Nevada (Rosenthal, 1969).

Long-term declines first slowed in the 1982–1985 period, when no change was observed from the previous 4 years. The reason(s) for this departure is not certain. The decline resumed into the period 1986–1989.

The most dramatic and unexpected finding in this report is the reversal after the late 1980s of decades-long declines in average Sr-90 concentration. We observed a 48.5% higher concentration in 1994–1997 births over 1986–1989 births (162 vs. 109 mBq Sr-90/g Ca), a trend consistent for each of five states (and counties in these states near nuclear reactors) included in this study. This temporal change cannot represent the continued decay of old bomb test fallout from Nevada; rather, it probably represents rising amounts of a currently produced source of environmental radioactivity entering the body. Current sources of Sr-90, a man-made fission product, are limited during the 1990s, and most are not likely to account for recently rising levels of Sr-90 in baby teeth.

(1) Fallout from the 1986 Chernobyl accident (including Sr-90) entered the US environment, raising levels of long-lived radionuclides, but these returned to pre-1986 levels within 3 years (Mangano, 1997; National Air and Radiation Environmental Laboratory, 1975–2001). For example, a rise of 98–311 mBq Cesium-137/l in pasteurized milk occurred in 60 US cities from May–June 1985 to May–June 1986, when Chernobyl fallout levels in the US peaked. This concentration in the same 2-month period in the following years

[5] Stamoulis et al. (1999) contains a chart summarizing trends in Sr-90 in deciduous teeth from various European nations and the Soviet Union. The chart shows that, from a level of approximately 10 mBq/g Ca in 1951, a peak of 250 was reached in 1964, similar to the US trend. By 1975, the average level had fallen to approximately 30 (three times the 1951 average) and was still declining. Three times the 1951 US average of just over 7 means that the 1975 US Sr-90 average should have been approximately 22. But the actual 1975 average found by RPHP was 183 (12 teeth), and 198 for 29 teeth from 1974–1976 births.

declined to 242, 155 and 81 mBq Cs-137/l, returning to pre-Chernobyl levels in 1989. Because Cs-137 has a half-life (30 years) similar to Sr-90 (28.7 years), it is logical that environmental (and thus, in vivo) Sr-90 from Chernobyl followed the same general pattern.

Another factor suggesting Chernobyl fallout probably does not account for the fact that post-1989 Sr-90 increases in baby teeth is the consistent finding of higher Sr-90 concentrations near nuclear power plants. Chernobyl fallout levels varied by geographic area, with the northwest US (where there is only one nuclear power reactor, in Washington state) receiving the highest level of radionuclide deposits.

(2) The increase probably does not represent high-level nuclear waste generated by reactors, which is generally stored in deep pools of cooled water or in casks below or above ground. Despite the leakage of some casks, the radioactivity contained in the waste is currently not in the food chain.

(3) Academic-based research reactors also produce fission products. However, these reactors are small in size and few (and declining) in number, which makes it an unlikely reason accounting for such a widespread and sustained trend in Sr-90 in bodies.

(4) Nuclear submarines produce fission products, but they are either contained within the submarine or released into the ocean. Thus, this is not a source of Sr-90 in the food chain, and not a reason for the rise documented in this report.

(5) Emissions from nuclear weapons plants account for another source of Sr-90. However, all reactors involved with producing nuclear weapons ceased manufacturing operations by 1991, and are not likely to play a role in rising Sr-90 concentrations after that time.

(6) While the last above-ground atomic bomb test took place in 1962, subterranean tests at the Nevada Test Site continued. Some of these tests vented radioactivity into the atmosphere. These emissions were much smaller than the atmospheric tests, and the last such test occurred in September 1992 (Norris and Cochran, 1994), making it an unlikely contributor to increases in Sr-90 throughout the 1990s.

(7) Reprocessing of nuclear fuels also creates fission products, but was ceased in the US in the late 1970s, and is not a factor in recent rises in Sr-90.

The only other source of Sr-90 that can explain this steady and dramatic rise in the 1990s is emissions from nuclear power reactors. Because reactors operated a greater percentage of the time, average annual generation of electricity rose 37.5% from 475 000 to 653 000 GW h from 1986–1989 vs. 1994–1997, an increase not markedly different from the 48.5% rise in average Sr-90 levels at birth (US Nuclear Regulatory Commission, 2001). Determining the extent of the correlation between these two trends requires more precise investigation.

Another major finding is that the counties located within 40 miles of each of six nuclear reactors have consistently higher Sr-90 levels than other counties in the same state. These counties were selected to generally correspond with those used by the US National Cancer Institute in a study of cancer near nuclear plants (Jablon et al., 1990). The excess near each nuclear plant ranged, with one exception, from 30.8 to 53.8% higher. More study, assessing whether locally produced radioactivity entering the body from inhalation and/or locally produced food and water account for these consistent differences, is merited. Findings on doses near reactors should be compared with health data. For example, childhood cancer rates near 14 of 14 eastern US reactors exceed the national rate (Mangano et al., 2003).

This analysis of proximity arrives at a different conclusion than an earlier report (O'Donnell et al., 1997) that found no correlation between distance from the Sellafield nuclear plant in western England and Sr-90 levels in baby teeth. That study used a regression equation to test this relationship. There are methodological and analytical differences between the two studies. O'Donnell considered teeth from as far as 300 miles from Sellafield, without taking into account Sr-90 produced by reactors other than Sellafield, while this report used only the counties most proximate (within 40 miles) to reactors. That report tested teeth in batches, while this study used individual readings. Factors other than distance from the radiation source may influence Sr-90 levels in vivo. The uptake of radioactivity in fetal tooth buds depends on intake during pregnancy/early infancy and transfer from maternal bone stores, which vary from person to person. These in turn can be dependent on food and water source, along with dietary differences.

A third major finding is that average Sr-90 concentrations vary geographically. Children from Pennsylvania (mostly near Pottstown, close to Philadelphia) who donated teeth had the highest average Sr-90 of the five states studied. Pottstown lies within 70 miles of 11 operating (and 2 closed) reactors, a concentration unmatched in the US. California, especially areas not close to nuclear reactors, is the state with the lowest average Sr-90. There are only four nuclear reactors on the entire west coast in operation since 1992, compared to dozens in the northeast.

At present (pending more detailed study), nuclear power reactors appear to be the most likely source explaining the recent unexpected rise in Sr-90 concentrations, and elevated Sr-90 levels nearest the plants. The geographic consistency and longevity of these trends and patterns, plus the large number of teeth studied, make these patterns meaningful and (in many instances) statistically significant. The fact that gross beta in US precipitation continued to rise after 1997 and that the highest average Sr-90 level since a low point was reached in 1986 occurred in the most recent birth year studied (1996, 195 mBq Sr-90/g Ca in 30 teeth) suggest that this trend may continue in the near future.

5. Study limitations/opportunities for further study

This report represents the first large-scale study of US in vivo levels of radioactivity in several decades. Although the initial findings presented here are important ones, they raise various questions that should be addressed in future research.

Other unexplored factors may help explain the temporal trends affected here. For example, the current study collected auxiliary data on mother's age at delivery and source of drinking water. Analyzing results by basic characteristics such as gender and race can be performed in future studies. Some factors that affect in vivo levels are already known. For example, children who are breast-fed accumulate lower Sr-90 concentrations than do bottle-fed infants (Rosenthal, 1969). Other dietary differences and their effects on Sr-90 levels can be further explored in future research.

Despite the consistency of results across geographic areas, substantial numbers of teeth were tested from only 5 of 50 US states. More teeth from other states would enhance knowledge about recent patterns of in vivo radioactivity. For example, 19 of the 50 US states (many in the western US) have no operating nuclear reactors, and may display patterns of Sr-90 different than the five already analyzed. The comparison could be extended to nations with no operating nuclear reactors (such as the Philippino teeth mentioned in this report). Testing the hypothesis that these states have lower levels of Sr-90 would be appropriate and necessary in future reporting of results.

The study did not collect sufficient teeth to compare local Sr-90 levels before and after a nuclear reactor opens. The hypothesis that opening a reactor will raise average in vivo concentrations and closing a reactor will reduce them should be tested.

A potential follow-up to this report is to institute a public program measuring in vivo levels in humans and/or animals near nuclear plants for the first time. In addition, more radionuclides in the environment (air, water, soil, etc.) may be tracked. The US government maintains such records near nuclear plants, but has phased out public reporting of several isotopes and failed to perform any long-term analysis.

The data presented herein describe past and current patterns of radioactivity in children's teeth. The three in vivo programs of measuring Sr-90 in US teeth and bones were never accompanied by any reports assessing potential health risks from this radioactivity. The current tooth study previously documented that average Sr-90 levels and childhood cancer rates followed similar trends during atmospheric bomb testing in the 1950s and 1960s. In addition, on Long Island, New York, recent Sr-90 trends correlate closely to trends in childhood cancer incidence, after a 3-year latency period (Gould et al., 2000a). Thus, comparing radioactivity and health patterns should be central to any follow-up of this analysis.

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Appendix A: Determination of Sr-90 to calcium ratio

Sr-90 in deciduous teeth was determined under the direction of Hari D. Sharma, Professor Emeritus of Radiochemistry and president of REMS, Inc., Waterloo, Ontario, Canada. Employing a 1220-003 Quantulus Ultra Low-Level Liquid Scintillation Spectrometer manufactured by the Perkin-Elmer Company in Massachusetts, Dr Sharma followed the following procedure.

Water-washed teeth were treated with 30% hydrogen peroxide for a period of 24 h to ensure that organic material adhering to teeth was oxidized. Teeth were then scrubbed with a hard brush for removing oxidized organic material and the fillings. Teeth were then dried at 110 °C for several minutes and then ground to a fine powder (ball mill). It is very important to remove any filling because if left behind inside a tooth, it tends to give colored solution or dissolution in a mineral acid. The presence of colored solution reduces the efficiency of counting.

Approximately 0.1 g of the powder is weighed in a vial, then digested for a few hours with 0.5 ml of concentrated nitric acid along with solutions containing 5 mg of Sr^{2+} and 2 mg of Y^{3+} carriers at approximately 110 °C on a sand bath. The solution is not evaporated to dryness. The digested powder is transferred to a centrifuge tube by rinsing with tritium-free water. Carbonates of Sr, Y and Ca are precipitated by addition of a saturated solution of sodium carbonate, and then centrifuged. The carbonates are repeatedly washed with a dilute solution of sodium carbonate to remove any coloration from the precipitate. The precipitate is dissolved in hydrochloric acid, and the pH is adjusted to 1.5–2 to make a volume of 2 ml, of which 0.1 ml is set aside for the determination of calcium. The remaining 1.9 ml is mixed with 9.1 ml of scintillation cocktail Ultima Gold AB, supplied by Packard Bioscience BV in a special vial for counting. A blank with appropriate amounts of Ca^{2+}, Sr^{2+} and Y^{3+} is prepared for recording the background.

The activity in the vial with the dissolved tooth is counted four times, 100 min each time, for a total of 400 min, with the scintillation spectrometer, to improve accuracy of results. The background count-rate in the 400–1000 channels is 2.25 ± 0.02 counts/min. The background has been counted for over 5000 min so that the error associated with the background measurement is approximately 1%. The overall uncertainty or one sigma associated with the measurement of Sr-90 per gram of calcium is ± 26 mBq/g Ca.

The efficiency of counting was established using a calibrated solution of Sr-90/Y-90 obtained from the National Institute of Standards and Technology, using the following procedure. The calibrated solution is diluted in water containing a few milligrams of Sr^{2+} solution, and the count-rate from an aliquot of the solution is recorded in channel numbers ranging from 400 to 1000 in order to determine the counting efficiency for the beta particles emitted by Sr-90 and Y-90. It is ensured that the Y-90 is in secular equilibrium with its parent Sr-90 in the solution. The counting efficiency was found to be 1.67 counts per decay of Sr-90 with 1.9 ml of Sr-90/Y-90 solution with 25 mg of Ca^{2+}, 5 mg of Sr^{2+}, 2 mg of Y^{3+} and 9.1 ml of the scintillation cocktail.

The calcium content was determined by using an Inductively Coupled Plasma instrument. The analysis is provided to REMS, Inc., by the University of Waterloo laboratories. REMS is located at P.O. Box 33030, Waterloo, Ontario, Canada, N2T2M9.

Appendix B: Calculation of counting error for Sr-90 in baby teeth due to laboratory observation and sample size

In Tables 3–7, the counting error for average concentrations of Sr-90 is calculated for each state as a combination of two variables: the error due to laboratory observation and the error due to sample size. Calculating each of these errors are as follows, using all 133 teeth (average mBq Sr-90/g Ca = 155) from Pennsylvania as an example. These data appear in Table 3.

Lab observation: The count of mBq of Sr-90 is not an exact one, but carries an uncertainty due to limitations of the counter. The error range for an individual tooth is ±26 mBq, a conservative estimate that may be lower for teeth with larger mass. Thus, the lab observation error for a sample of 133 teeth is

$$26\ \text{mBq}/\sqrt{N} = 26\ \text{mBq}/\sqrt{133} = 2.25\ \text{mBq}$$

Sample size: The error due to the sample size is

$$1/\sqrt{N} = 1/\sqrt{133} = 13.44\ \text{mBq}$$

Calculation: The squares of the two results are added quadratically. Thus,

$$\sqrt{((2.25)^2 + (13.44)^2)} = 13.63\ \text{mBq (rounded to 14)}$$

With an average Sr-90 concentration for the 133 teeth of 155 mBq/g Ca, the confidence interval is between 127 and 183, or 155 plus or minus 28 (2 times 14). Thus, there is a 95% chance that the actual average of the entire population falls within 127 and 183.

Appendix C: Calculation of significance of differences in Sr-90 averages between counties near reactors and more distant counties

In Table 4, average Sr-90 concentrations in teeth from counties near nuclear reactors were compared with averages from other counties in the same state. The significance of differences between the two means was calculated using a *t*-test.

For example, the mean Sr-90 concentration for counties closest to the Indian Point reactor was 164 mBq/g Ca (217 teeth), compared to 121 (317 teeth) for other counties in New York State. The formula used for the significance of this difference is as follows:

Counties near Indian point: $\pm\{1/\sqrt{217}\} \times 164 = 11.1$ (rounded to 11)

Other counties in New York state: $\pm\{1/\sqrt{317}\} \times 121 = 6.8$ (rounded to 7)

$$\{164 - 121\}/\sqrt{(11^2 + 7^2)} = (45/13.04) = 3.45$$

In a basic statistics table, 3.45 standard deviations (z score) indicate a P value of <0.001, i.e. there is less than a 1 in 1000 chance that the difference is due to random chance.

In Tables 5–7, the significance of differences in average Sr-90 concentrations from 1986–1989 to 1994–1997 were tested using a similar technique. For example, using Florida data in Table 6

1986 − 1989; for 102 teeth, average mBq Sr-90/g Ca = 112

1994 − 1997; for 99 teeth, average mBq Sr-90/g Ca = 153

$1986-1989 = \pm\left\{1/\sqrt{102}\right\} \times 112 = 11.1$ (rounded to 11)

$1994-1997 = \pm\left\{1/\sqrt{99}\right\} \times 153 = 15.4$ (rounded to 15)

$$\{153 - 112\}/\sqrt{(11^2 + 15^2)} = 2.20$$

In a basic statistics table, 2.20 standard deviations (z score) indicate a P value of <0.04, i.e. there is less than a 4 in 100 chance that the difference is due to random chance.

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