Time Trends of Persistent Organic Pollutants and Heavy Metals in Umbilical Cord Blood of Inuit Infants Born in Nunavik (Québec, Canada) between 1994 and 2001

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Inuit inhabitants of Nunavik (northern Québec, Canada) consume great quantities of marine food and are therefore exposed to high doses of food chain contaminants. In this study, we report the time trends of persistent organic pollutants, mercury, and lead in umbilical cord blood of infants from three communities of the east coast of Hudson Bay in Nunavik. We analyzed 251 cord blood samples collected from 1994 through 2001 for polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethane (DDT), dichlorodiphenyl dichloroethylene (DDE), hexachlorobenzene (HCB), chlordanes, lead, and mercury. Using an exponential model, we found strongly significant decreasing trends for PCBs (7.9% per year, p < 0.001), DDE (9.1% per year, p < 0.001), DDT (8.2% per year, p < 0.001), and HCB (6.6% per year, p < 0.01). No significant trends were detected for chlordanes. A significant reduction of lead and mercury concentrations was found, but there was no clear linear or exponential trend. The decreases observed could be explained by a decrease in food contamination, by changes in dietary habits, or, most likely, by a combination of both. Key words: aboriginal, chlorinated pesticides, diet, environmental exposure, food contamination, heavy metals, human, lead, mercury, newborn, polychlorinated biphenyls, time trend, umbilical cord blood. Environ Health Perspect 111:1660-1664 (2003). doi:10.1289/ehp.6269 available via http://dx.doi.org/ [Online 2 July 2003]

Lipophilic organochlorines (OCs) that resist biodegradation can accumulate in the environment to become persistent organic pollutants (POPs). Among them, polychlorinated biphenyls (PCBs) and several chlorinated pesticides have been detected in tissues of animals and human throughout the world. Their capacity to accumulate in adipose tissue leads to biomagnification in the food chain, and their concentrations reach highest levels in top predator species (Braune et al. 1999; Muir et al. 1999). Mercury and lead are ubiquitous in the environment. They both occur naturally, but human activity has increased their mobilization and distribution in the environment. Mercury is excreted slowly by animals and plants and also accumulates in the food chain (found mostly as methylmercury, its organic form).

Most studies focusing on temporal trends of POPs have identified a decreasing trend during the last decades. Since the mid-1970s, levels of dichlorodiphenyl trichlorethane (DDT) and PCBs have decreased in tissues of freshwater fishes and herring gull eggs in Canada and the United States (Hebert et al. 1994; Ryckman et al. 1994; Schmitt et al. 1999). In Arctic wildlife, POP concentrations seem on the decline for most species (Muir and Norstrom 2000; Muir et al. 1999) but not all (Muir et al. 2000). In environmentally exposed humans, levels of PCBs in breast milk have dropped in the last 15 years in Germany (Schade and Heinzow 1998) and Sweden (Noren 1993; Noren and Meironyte 2000). Downward trends in human fluids have also

been observed in the United States (Michigan; He et al. 2001), Mexico (Waliszewski et al. 1998), Canada (Dallaire et al. 2002), and the United Kingdom (Harris et al. 1999).

For cultural and economic reasons, carnivorous fish and marine mammals constitute an important part of the diet of the Inuit population living in Nunavik (northern Québec, Canada). Their exposure to such biomagnified substances as OCs and heavy metals is thus proportionally high. Several studies have identified markedly higher mean concentrations of POPs and heavy metals in adult blood, cord blood, and breast milk of Nunavik inhabitants compared with those of the southern population of the province of Québec (Ayotte et al. 1997; Dewailly et al. 1989, 1993, 1998; Muckle et al. 1998; Rhainds et al. 1999). In this context, we report here the temporal variations of POPs, mercury, and lead in umbilical cord blood of infants born from 1994 through 2001 in three communities of the coast of the Hudson Bay in Nunavik.

Materials and Methods

Population and recruitment. The targeted participants were pregnant Inuit women living in three communities (Puvirnituq, Inukjuaq, and Kuujjuaraapik) on the east coast of Hudson Bay in Nunavik (Figure 1). For the present analysis, we included the participants of two previous studies done on the same population. The first study was designed to monitor the prenatal exposure to environmental contaminants and took place between November 1993 and December 1996 (Dewailly et al. 1998). Pregnant Inuit women were invited to participate on arrival at the health center for delivery. Of the 491 women who accepted the invitation to participate, only the women living in the three targeted communities (Puvirnituq, Inukjuaq, or Kuujjuaraapik) were included in the present analysis (n = 138). We excluded women living in other communities because we had a significant number of samples only for 3 years for these communities, and we believed that the interference caused by annual variations would have been too great for the estimation of valid time trends.

The second study was designed to evaluate the impact of environmental exposure to POPs and heavy metals on infant health and development (Muckle et al. 2001). It was conducted between November 1995 and March 2002 in the three communities mentioned above. In this study, pregnant women were approached by one of our research assistants after their first prenatal medical visit. Three hundred fifty-eight eligible pregnant women were approached, and 248 (69.3%) accepted the invitation to participate. Because of staff shortage during deliveries, only 113 cord blood samples were available when statistical analyses were done. The participants for whom the cord blood samples were unavailable were similar to the other participants regarding gestational age, parity, maternal age, and infant sex. However, they had slightly lower maternal blood mean concentration of OCs. The 113 participants for whom cord

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The authors declare they have no conflict of interest. Received 10 February 2003; accepted 2 July 2003. blood was available and the 138 participants of the first study composed our study population for the present analysis (n = 251).

All women who agreed to participate in either study signed an informed consent form and could withdraw at any time. Both protocols were approved by the Laval University ethics committee.

Data collection. The data collection and all the laboratory procedures were rigorously the same for both studies. We sampled blood (10-20 mL) from the umbilical cord at delivery immediately after it was severed. An aliquot of blood was centrifuged, and the plasma was transferred into a glass vial prewashed with hexane. Blood and plasma were kept frozen at -80°C and sent to the Institut National de Santé Publique du Québec (Québec City, Canada) every 3 or 4 months for contaminant and biochemical analyses. We gathered information on parity, maternal age, and infant sex, gestational age, and birth weight from the medical charts of the mother and the newborn a few weeks after delivery.

Determination of OCs. Details on the procedures used by our laboratory, as well as information on the quality control of our chemical analyses, have been described previously (Rhainds et al. 1999). We determined the concentrations of 14 PCBs congeners (International Union for Pure and Applied Chemistry numbers 28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, and 187) and of 11 chlorinated pesticides [aldrine, α -chlordane, γ -chlordane, *cis*-nonachlor, *p*,*p'*-DDE (dichlorodiphenyl dichloroethylene), p,p'-DDT, hexachlorobenzene (HCB), mirex, oxychlordane, trans-nonachlor, and β-hexachlorocyclohexane] in plasma samples by high-resolution gas chromatography. Plasma samples (2 mL) were extracted, cleaned on Florisil columns, taken to a final



Figure 1. Geographic location of Nunavik, Québec, Canada.

volume of 100 μ L, and analyzed on an HP-5890 series II gas chromatograph equipped with dual-capillary columns and dual ⁶³Ni electron-capture detectors (Hewlett-Packard, Palo Alto, CA, USA). We identified peaks by their relative retention times obtained on the two columns using a computer program developed in-house. The limit of detection was 0.02 μ g/L for all OCs analyzed. Because OCs are stored mainly in body fat, the concentrations are expressed on a lipid basis.

Determination of blood lipids. We measured total cholesterol, free cholesterol, and triglycerides in plasma samples by standard enzymatic procedures. Phospholipid concentrations were determined according to the enzymatic method of Takayama et al. (1977) using a commercial kit (Wako Pure Chemical Industries, Richmond, VA, USA). We estimated the concentrations of total plasma lipids using the formula developed by Phillips et al. (1989).

Determination of heavy metals. We determined total mercury concentrations by coldvapor atomic absorption spectrometry. Samples were digested with nitric acid, and mercury was reduced by adding anhydrous stannous chloride and cadmium chloride. Metallic mercury was volatilized and detected by atomic absorption spectrometry (model 120; Pharmacia, Piscataway, NJ, USA). The detection limit for blood mercury analysis was 1.0 nmol/L. We determined lead concentration by diluting an aliquot of whole blood with a mixture of nitric acid, ammonium phosphate, and Triton X-100. We then analyzed it by graphite furnace atomic absorption with Zeeman background correction (model ZL 4100; Perkin Elmer, Norwalk, CT, USA). The detection limit of the method was 50 nmol/L.

Determination of fatty acids in plasma phospholipids. For the determination of the fatty acid composition in plasma phospholipids, 200-µL aliquots of plasma were extracted after the addition of chloroform:methanol (2:1, vol/vol) in the presence of a known amount of internal standard (diheptadecanoyl phospholipid). We isolated total phospholipids from the lipid extract by thin-layer chromatography using heptane:isopropyl ether:acetic acid (60:40:3, vol/vol/vol) as the developing solvent. Following transmethylation, with use of BF3/methanol, the fatty acid profile was determined by capillary gas-liquid chromatography. The fatty acid composition of plasma phospholipids was expressed as percentages of the total area of all fatty acid peaks from C14:0 to C24:1 (Holub et al. 1987).

Statistical analysis. We assigned a value of half the detection limit of the analytic method when a compound was not detected in a sample. In all statistical analysis, we considered only those substances for which 50% of the samples were above the limit of detection. This was done because trends calculated from a majority of samples with imprecise values (below the limit of detection) would most likely yield biased results. Contaminant concentration variables had log-normal distributions and were log-transformed for all analyses.



Figure 2. Adjusted mean OC concentrations according to the year of birth: (*A*) PCBs; (*B*) DDE; (*C*) HCB; (*D*) oxychlordane. The lines represent the slope estimates presented in Table 2.

Therefore, all contaminants results are presented as geometric means. All the other continuous and discrete variables are presented as arithmetic means ± standard deviation. For temporal trends, we performed multiple regression modeling using the year of birth of the baby as the main independent variable. Because the dependent variables (contaminant concentrations) of the regressions were logarithmically transformed, the estimated slope of the regression (β) can be interpreted as the number of log increase (or decrease) of contaminant concentrations per year. From these results, we calculated the percentage of increase (or decrease) of contaminant concentrations per year using the expression $[(1 - e^{\beta}) \times 100]$. To identify potential confounders, we selected variables that were associated with one or more contaminants and that were not constant in time. We did not consider dietary variables because our hypothesis included dietary changes as a potential explanation for temporal variations. We controlled for the village of residence, maternal age (continuous), parity (continuous), and the season of birth. We excluded birth weight and gestational age because these variables did not affect the association between contaminants and the year of birth. We also estimated adjusted means (least-square means) for each year using multiple regression in order to produce Figures 2 and 3. In this case, contaminant concentrations in cord blood of infants born in 2000 and 2001 were merged because



Figure 3. Adjusted mean heavy metal concentrations according to the year of birth for (*A*) lead and (*B*) mercury. The solid lines represent the slope estimates presented in Table 2. The dotted lines represent the adjusted mean lead concentrations before and after January 1999.

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numbers were small in 2001 (n = 5). The database management and all the statistical analyses were performed with SAS software (version 8.0; SAS Institute, Cary, NC, USA). By convention, we considered p < 0.05 statistically significant.

Results

After merging of the data from the two studies, 251 participants could be included in the analysis. Concentrations of OCs, mercury, and lead were available for 238, 240, and 242 participants, respectively. Table 1 shows the descriptive characteristics of the participants. The mean age at delivery was 23.5 ± 4.5 years. About half of the participants were from Puvirnituq, 36.3% were from Inukjuaq, and the remaining 11.1% were from Kuujjuaraapik (Table 1). Consistent with previously published data, maternal age and parity were strongly associated with most OCs in cord blood (data not shown).

Table 2 presents the unadjusted and adjusted annual decreases for contaminant concentrations when an exponential model is used. Except for heavy metals, the adjusted model yielded slightly steeper slopes and smaller *p*-values than did the unadjusted model. The concentration of the sum of PCB congeners decreased by 6.1% per year in the unadjusted model (p < 0.05) and by 8.5% per year in the adjusted model (p < 0.001). For individual PCB congeners, all trends were statistically significant, except for PCB 153 and PCB 170 in the unadjusted model.

For chlorinated pesticides, we detected significant declining trends in the concentrations of p,p'-DDE and p,p'-DDT. The decrease for p,p'-DDE concentration was 9.3% per year in the adjusted model (p < 0.001). The ratio of DDT to DDE concentrations was stable across the entire study period (data not shown). HCB also decreased significantly by 4.8% (unadjusted) and 6.4% (adjusted). We could not detect any significant trends in chlordane concentrations.

Both mercury and lead concentrations decreased by more than 8% per year for the

 Table 1. Descriptive characteristics of the participants.

Characteristics	Arithmetic mean	Percent	
Mothers			
Age in years (n = 247)	23.5 ± 4.5		
Number of previous			
pregnancies (n = 248)	2.1 ± 1.8		
Residence (<i>n</i> = 251)			
Puvirnituq		52.6	
Inukjuaq		36.3	
Kuujjuaraapik		11.1	
Newborns			
Sex (% male) (<i>n</i> = 249)		52.2	
Gestational age			
(weeks; n = 213)	39.2 ± 1.4		
Birth weight			
(grams; <i>n</i> = 245)	3,513 ± 438		

study period. The trends were strongly significant both for the unadjusted and for the adjusted models.

Figures 2 and 3 illustrate the temporal trends for OCs (Figure 2) and heavy metals (Figure 3). The solid lines represent the adjusted trends as presented in Table 2. It is interesting to note that, although the exponential model for lead was significant, visual inspection of the data did not support an exponential decrease (Figure 3). The mean concentrations were fairly constant until 1998 and sharply dropped in 1999. In fact, the fit of a statistical model contrasting the mean levels before and after 1999 (represented by the dotted lines) was better than the fit of the exponential model (solid line). Figure 3 also shows that mercury levels seemed constant in time, except in 1998 and 2000, despite what was found using an exponential model.

We examined n-3 fatty acid proportions as a surrogate of maternal fish consumption (Silverman et al. 1990) to see if a diminution of fish consumption had occurred during the study period. A slight decrease in n-3 fatty acids was detected, but it was not statistically significant. Expressed as a percentage of the total fatty acids, mean n-3 fatty acid proportion was 4.8% in 1994, and the model yielded a decrease of 0.06% per year (p < 0.28).

Discussion

In this study, we identified an exponential decrease in most of the contaminants analyzed. Knowing that the major source of exposure in our study population is the ingestion of traditional food items of high trophic level, the decrease observed could be explained by a decrease in food contamination, by changes in dietary habits, or by a combination of both.

In Canada, the use of chlorinated pesticides and PCBs has been severely restricted since the 1970s. The use of most chlorinated pesticides is banned, and only closed-use PCBs in already existing equipment are still allowed. Today, it is believed that local sources of PCBs, mainly from abandoned contaminated arctic military sites, do not contribute significantly to human exposures in the Arctic. The comparison of PCB congener signature in soil showed that the impact of arctic contaminated sites was limited to their immediate vicinity, a halo of a few kilometers (Bright et al. 1995). The situation is similar for chlorinated pesticides. They have been used in the 1940s and the 1950s for insect control in Nunavik but are unlikely to be used today because of regulations (although information on the use of pesticides in the Arctic is scarce).

Nevertheless, release in the environment still occurs because of storage leakage and ongoing use in certain parts of the world. OC contaminants are distributed throughout the globe and reach arctic regions by long-range atmospheric transport and oceanic currents (Barrie et al. 1992; Burkow and Kallenborn 2000; Thomas et al. 1992). They accumulate in the food chain, and it is now well accepted that the high trophic level of the traditional Inuit diet is mainly responsible for the high exposure of Inuit populations to biomagnified substances such as POPs (Bjerregaard et al. 2001; Dewailly et al. 1993; Kuhnlein et al. 1995).

The eating habits of Inuit populations have changed enormously during the last 50 years. Since market-bought food has been introduced in their diet, added carbohydrates, junk food, pork, chicken, milk products, and other "foreign" food items have become increasingly popular, especially among adolescents and young adults (Blanchet et al. 2000; Moffatt et al. 1994; Murphy et al. 1995; Olsen 1985). Market food usually has a lower trophic level than does traditional Inuit food and is consequently less contaminated by POPs. This is reflected by the much lower mean concentrations of PCBs and chlorinated pesticides detected in cord blood samples from populations whose diet is almost exclusively composed of market-bought food, such as those in southern Québec (Rhainds et al. 1999). However, although a gradual switch from traditional food to market food would result in a decrease in blood concentrations of food chain contaminants, it seems unlikely that dietary modifications would be of such magnitude that they alone would cause an annual decrease of 5-10% in the body burden of contaminants.

Information on OC time trends in wildlife is scarce. Since the 1980s, there seems to be a decline in tissue concentrations of arctic marine and terrestrial mammals (Muir et al.

1999; Muir and Norstrom 2000). Scattered reports also underline a general decrease in OCs for various species in different regions of the world: cod, flounder, mussels, and shrimp (Roose et al. 1998), foxes (Georgii et al. 1994), freshwater fishes (Schmitt et al. 1999), and herring gulls (Hebert et al. 1994; Ryckman et al. 1994). In environmentally exposed humans, almost all studies addressing the question of temporal trend of OCs have found a decreasing tendency (Dallaire et al. 2002; Harris et al. 1999; He et al. 2001; Noren 1993; Noren and Meironyte 2000; Schade and Heinzow 1998; Waliszewski et al. 1998). In these studies, the time required to halve the mean contaminant concentrations in the population $(t_{1/2})$ for PCB and DDT/DDE ranged from 4 to 7 years, except for PCBs in breast milk of Swedish women, which had a $t_{1/2}$ of 14 years (Noren and Meironyte 2000). These results are slightly lower but remain similar to what we observed in the present study ($t_{1/2}$ = 7.8 years and 7.1 years for PCBs and DDE, respectively). The generalized downward tendency of OC concentrations observed in wildlife and human tissues throughout the world strongly suggests that the environmental contaminant burden is steadily declining and that this tendency can be observed in all levels of the food chain. We believe that most of the decrease of OC concentrations observed in this study can be attributed to descending concentrations in the traditional food items of the Inuit diet.

Sources of mercury and lead are diverse and are from both natural and anthropogenic origins. They are taken up by Arctic biota and depend on both local human activities and long-range transport (Dietz et al. 1998). Mercury and its organic form, methylmercury,

Table 2. Annual	percentage of	decrease ^a of	contaminants	concentration in	umbilical cord blood

		Annual decrease in percentage (95% CI)		
Contaminants	Percent detected ^b	Unadjusted ($n = 238$) ^c	Adjusted ^d $(n = 234)^e$	
PCBs and pesticides				
Σ PCB congeners		6.1 (1.0–10.9)*	8.5 (3.7–13.1)#	
PCB-99	94.6	7.9 (3.1–12.5)**	9.5 (4.7–14.1)#	
PCB-118	90.5	7.1 (2.3–11.7)**	9.1 (4.4–13.6)#	
PCB-138	100.0	5.7 (0.5–10.7)*	8.2 (3.2-13.0)**	
PCB-153	100.0	5.1 (-0.4-10.4)	7.6 (2.2–12.7)**	
PCB-170	80.2	4.5 (-1.6-10.2)	7.5 (1.9–12.8)**	
PCB-180	99.2	8.1 (2.5–13.3)**	11.0 (5.9–15.8)##	
PCB-187	95.4	5.4 (0.6–9.9)*	8.0 (3.7–12.1)#	
DDE	100.0	6.7 (6.4–11.5)*	9.3 (4.4–14.0)#	
DDT	71.5	5.9 (1.1–10.4)*	8.1 (3.6–12.2)#	
HCB	100.0	4.8 (0.3–9.0)*	6.4 (2.1–10.6)**	
<i>cis</i> -Nonachlor	74.0	0.4 (-4.9-5.4)	0.9 (-4.4-5.8)	
Oxychlordane	94.6	0.9 (~5.4–6.7)	2.8 (-3.5-8.6)	
trans-Nonachlor	100.0	2.3 (~2.8–7.1)	4.0 (-1.0-8.8)	
Heavy metals				
Pb	100.0	8.2 (3.5–12.5)#	8.7 (4.5–12.7)##	
Hg	100.0	9.4 (5.0–13.6)##	8.3 (3.8–12.7)#	

^{*a*}Percentage decrease per year calculated by multiple regression in which the year of birth is a continuous variable. Because the dependant variable (contaminants concentrations) of the regression was logarithmically transformed, each annual decrease and 95% CI were calculated from the slope (β) of the regression estimate (and its 95% CI) according to $(1 - e^{\beta}) \times 100$). ^{*b*}Percentage of samples for which the contaminant was above the limit of detection. ^{*c*}*n* = 242 for lead; *n* = 240 for mercury. ^{*d*}Adjusted for the village of residence, maternal age, parity, and season of birth. ^{*e*}*n* = 237 for lead; *n* = 235 for mercury. ^{*s*}*p* < 0.001; ^{*f*}*p* < 0.001.

bioaccumulate in the food chain reaching the highest level in the meat of sea mammals. Again, it is mainly through their traditional diet that Inuit are exposed to mercury (Bjerregaard and Hansen 2000; Dewailly et al. 2001; Johansen et al. 2000; Muckle et al. 2001). Recent data on temporal trends of mercury burden in wild animals are very limited. Wagemann et al. (1996) have noted an increase in mercury concentrations in beluga, ringed seal, and narwhal of the Canadian arctic between 1981 and 1994. In environmentally exposed humans, no statistically significant trends were noted in hair in a population of Seychelles islanders between 1986 and 1989 (Cernichiari et al. 1995). Because no data on mercury temporal trends in arctic wildlife since 1994 are available, it is hazardous to speculate on the cause of the variations observed in this study. In contrast to OCs, our results for mercury do not support an exponential decrease. The concentrations were constant across the years, except for markedly lower levels in 1998 and 2000. When we used n-3 fatty acids in cord blood as a surrogate of maternal fish consumption, we observed a slight decrease that was not statistically significant and was not related to the mercury concentration. We also searched for relation between mercury concentration and the numbers of beluga caught in each village between 1996 and 2001, but no relation was found (Department of Fisheries and Oceans 2002). A thorough dietary survey would be necessary in order to elucidate the cause of the variation observed. Other studies with longer follow-up would clarify whether the lower levels observed in 1998 and 2002 were due to chance or were signs of a new temporal trend of mercury.

The sources of exposure to lead are less clear. Biomagnification of lead in the food chain does not play such an important role (Dietz et al. 1998; Muir et al. 1992). Rather, a recent study in Nunavik concluded that an important part of lead exposure could be from lead shots used for hunting (Lévesque et al. In press). Many reports have addressed the question of temporal trends of lead in human blood. Most authors have noted a strong decrease since the 1980s, mainly because of the switch from leaded to unleaded gasoline (Benes et al. 2001; Ducoffre et al. 1990; Kalina et al. 1999; Rothenberg et al. 2000; Wietlisbach et al. 1995). Few data are available on lead trends in tissues of arctic wildlife species. However, environmental lead contamination is low in the arctic biota and does not seem to be the primary source of exposure for Inuit population (Lévesque et al. In press). Unlike OCs and mercury, the observed decrease in lead concentration seems to be caused by the recent ban of lead shot combined with an information campaign on the potential health effects of lead exposure due to lead shots. Our data strongly support this hypothesis: Lead concentration fell markedly in 1999, when lead shots were banned (0.12 μ mol/L after the ban vs. 0.20 μ mol/L before the ban, p < 0.0001). In fact, the fit of the statistical model is better when lead geometric mean levels before and after the ban are compared (in contrast to the exponential models used throughout this study for the other contaminants).

The present analysis was based on the merging of results from two different studies. We did not collect information from mothers refusing to participate in either study, so we cannot ascertain whether a selection bias was introduced in our results. Shortage of staff for cord blood sampling during delivery may have also introduced such a bias because participants for whom cord blood was available had slightly higher mean levels of OCs in their blood (maternal blood sample). The trends may therefore be underestimated. Finally, women with complicated or at-risk deliveries were usually transferred to other hospitals. Our results therefore overrepresent healthier and uncomplicated pregnancies and deliveries.

Since the 1970s, many restrictions and regulations have helped drastically reduce the input of POPs and heavy metals in the environment, and exposure through food contamination decreased accordingly. In our study population, we believe that POP decline is caused mainly by a diminution of food contamination and, to a lesser extent, dietary changes. It was well beyond the scope of this study to try to determine the specific contribution of these two factors to the observed decrease. Although questions remain as to the exact causes of the decline, it is encouraging to observe such an improvement in prenatal exposure for this highly exposed population. International efforts to further reduce the environmental input should be continued.

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