EXPOSURE OF FISHING PEOPLE TO CHLORINATED AROMATIC HYDROCARBONS IN THE LOWER NORTH SHORE OF THE ST. LAWRENCE RIVER, QUÉBEC, CANADA

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The lower north shore of the St. Lawrence River in Québec comprises a number of small settlements which are remote from the rest of the province being some 2000 km northeast of Montréal. These maritime people of Caucasian origin many of whom are fishermen by occupation depend on wild animals not only for their revenue but also as a major source of food. As a result their consumption of food in the form of finfish, crustaceans, marine mammals, terrestrial game, and seabirds is considerable[1]. Data on the chlorinated aromatic hydrocarbon content of animals from this region in the gulf of the St. Lawrence is scanty[2,3] but indicates that it is similar to other areas such as the Great Lakes. Thus high levels of consumption of wildlife from this region may result in elevated exposure to contaminants such as the PCBs and PCDDs/PCDFs. For this reason we investigated the environmental exposure of these people by determining concentrations of these contaminants in blood plasma.

METHODS

Of the more than 4300 people in the region, those individuals and their family members over 15 yr and residing in the area for at least 10 yr were randomly selected from the registries of the fishing associations of the major settlements. From the 422 individuals requested in March/April of 1990, 201 fasting blood plasma samples of about 40 mL were obtained for which analysis were carried out for the PCBs, organochlorines, heavy metals, liver enzymes, blood lipids, and fatty acids. An additional 25 blood samples for the determination of the organochlorines only were performed on samples taken in February 1991 from those individuals in two villages with the highest total PCB concentrations. All donors filled out a detailed questionaire on their dietary habits including the amount consumed of fish, seafood, birds and their eggs, and terrestrial game.

Determination of PCB congeners was carried out on about 2 mL of plasma by saponification, extraction with hexane-ether, chromatography on 2% water deactivated Florisil with hexane elution, and measurement by gas chromatography (GC) on a DB-5 non-polar silicone column with electron capture detection. Quantification was carried out on the 11 most prominent congeners (IUPAC numbers by order of GC elution: 28, 52, 101, 118, 153, 138, 156, 187, 183, 180, 170) using a standard curve with a limit of detection per congener of about 0.2 g/L.

Determination of the co-planar PCBs (#s 126 and 169) and the PCDDs/PCDFs with four or more chlorines and 2,3,7,8-substitution (usually about 12 congeners) was accomplished on about 30 mL of plasma using isotope dilution mass spectrometry (MS). Isotope labelled $^{13}C_{12}$PCDDs, $^{37}Cl_4$PCDFs, and $^{13}C_{12}$PCBs were added to the plasma, followed by ethanol, and ammonium sulfate, and the mixture extracted twice with hexane. After weighing of the evaporated total hexane extracts to constant weight for the lipid content, the extracts in hexane were defatted with H_2SO_4, purified on columns of acid/base silica, Florisil and carbon, and measured by GC-MS on a non-polar DB-5 column as outlined previously[4]. Both co-planar PCBs and PCDDs/PCDFs were present in the same final extract. Their quantification was carried out by separate injections on the MS using a standard curve, and relative response factors. With this sample size, the detection limit of the individual congeners on a blood lipid basis was about 10 ng/kg (parts per trillion; ppt).

RESULTS AND DISCUSSION

The average total PCB plasma concentration on a wet weight basis for 185 of the individuals was 12.3μg/kg. The major contributors to this total were the hexa- congeners # 138 (18%) and # 153 (27%) and the hepta- congener (20%). Congeners # 28, 52, and 101 were virtually absent. Two of the congeners (# 118 and # 156) are mono-*ortho* chloro substituted and were detected in most samples (# 118 at 6% of total). The ten individuals with the highest total PCB levels overall averaged 46.4μg/kg and the highest 25 samples collected in 1991 from two villages showed average levels of 26.3μg/kg. Comparison of these total PCB concentrations to the values of about 1 to 1.5μg/kg found in pooled plasma samples collected from individuals residing in the more populous parts of Québec and Ontario shows a difference of at least an order of magnitude (factor of 10 times).

Concentrations of the co-planar PCBs and PCDDs/PCDFs were measured only in the 25 samples collected in 1991. The levels of the co-planar PCB congeners were much lower than those reported for the other PCBs i.e. less than 0.1% of total PCBs. On a lipid basis, congener # 126 (penta-) averaged 620 ng/kg and congener # 169 (hexa-) showed an average value of 580 ng/kg. A pooled sample of blood plasma collected in Ontario from Red Cross donors in 1988 showed concentrations for the same two congeners of about 30 to 40 ng/kg i.e. a least an order of magnitude lower than the fishing population under study. Concentrations of those PCDDs/PCDFs often associated with fish consumption/exposure, 2,3,7,8-TCDD and 2,3,4,7,8-PnCDF, were 14 and 33

ng/kg, respectively. These levels are about three times higher than a control population of plasma sampled from Montréal in 1988[5]. However on a TCDD toxic equivalency basis (TEQ) using all measured congeners, the difference between the fishing and control group was only a factor of two.

CONCLUSION

Fishing people from the lower north shore of the gulf of St. Lawrence, Québec have levels of total PCBs, and co-planar PCBs at least ten times higher than those from other parts of the province. The same comparison for the PCDDs/PCDFs shows a smaller difference in the range of 2 to 3. Some of this difference could be due to age of the donors. On a TEQ basis, the PCDDs/PCDFs, co-planar PCBs, mono *ortho* PCBs, and other PCBs contribute to the total dioxin-like toxicity[6] in proportions of 5%, 35%, 50%, and 10%, respectively. The source of these elevated contaminants in blood is not certain but data from questionaires indicate wildlife consumption particularly birds' eggs plays a major role. A more detailed assessment of the levels of these contaminants in the foods consumed in this area is presently under investigation.

ACKNOWLEDGEMENTS

The authors are thankful for the contributions of the health personnel of the lower north shore and Centre Hosp. Univ. Laval, Québec for sample collection, to J.-P. Weber and group of Centre Toxicologie, Univ. Laval for PCB determinations, and to L. Panopio, W.-F. Sun, and M. Boyle of Health Protection Branch, Ottawa for co-planar PCB and PCDD/PCDF determinations.

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