Inuit Greenland Exposure to Dioxin-like Compounds

John J Ryan
Bureau of Chemical Safety, Health Canada, Ottawa K1A 0L2, Canada
Eric Dewailly and Pierre Ayotte
Public Health Centre of Quebec, 2400 d'Estimauville, Beauport, Quebec G1H 7G9, Canada
Henning S. Pedersen and Gert Mulvad
Center of Primary Care, P O Box 1001, DK-3900, Nuuk, Greenland/Denmark
J. C. Hansen
Aarhus University, Bldg 180, Universitetsparken, DK-8000, Aarhus C, Denmark

1. Introduction

Much of the production and use of persistent anthropogenic compounds takes place in industrialized areas in temperate climates. Many of these chemicals are somewhat volatile, become airborne and, by simple distillation, are transported from warmer to cooler climates[1,2]. Because of their tendency to associate with lipids, these compounds then accumulate in animal species residing in areas remote from the origin of the contaminant. Chlorinated aromatic hydrocarbons appear to follow this scenario as significant concentrations are now found in aquatic animals at the top of the food chain in polar regions. Consumption of these animals by indigenous human populations can lead to important exposure to dioxins, furans, PCBs, and other tenacious organochlorines. For example, Inuit mothers living in northern Quebec have milk levels of total PCBs[3] and other OCs[4] from two to more than ten times that of residents from the more populated regions in the south. Similar differences have been noted between individuals in the northern and in the populated areas for non *ortho* (coplanar) and mono *ortho* PCBs[5] again using milk analyses. When this exposure assessment is expanded to both sexes and all ages by using blood plasma as index, the differences for the PCBs and dioxin-like compounds are still between five and fifteen times[6].

Information on the body burden of other peoples living in northern or remote areas is rather limited. Recently we reported on the total and some congener PCB content of autopsy tissues collected in Nuuk, Greenland from Inuit who resided in Greenland[7]. Average concentrations in omental fat for the sum of fourteen PCB congeners was over 6 mg/kg on a lipid basis with the major congeners being PCB numbers 138, 153, and 180. These human values were some of the highest reported to date and appeared to be more than ten fold greater than levels from other countries. We now describe the dioxin-like compounds in both omental fat and liver from some of these same tissues. Dioxin-like compounds are here used to include seventeen 2,3,7,8-substituted PCDDs and PCDFs[8] and certain PCB congeners[9] including three non *ortho*, eight mono *ortho* and two di *ortho* for a total of 30 compounds.

2. Method

Sampling Autopsy tissues from individuals of approximately equal numbers of both sexes, average age about 57 yr, who died from various causes have been described[7]. Of the four tissue types collected, omental and subcutaneous adipose, liver, and brain, paired samples of omental fat and liver from 26 individuals and omental fat only from 15 individuals (total of 41 individuals and 67 samples) were present in large enough quantities for dioxin-like determinations. The results from thirteen persons (26 paired tissues) are given here.

Analysis After tissues had been extracted, defatted, and separated into two fractions by chromatographic adsorbents, the seventeen 2,3,7,8-substituted congeners of the PCDDs/PCDFs and the three non *ortho* PCBs (total 20 analytes) were measured by gas chromatography (GC)-mass spectrometry (MS) and the eight mono *ortho* and two di *ortho* PCBs (ten congeners) by GC-electron capture detection. In summary, a mixture of $^{13}C_{12}$-2,3,7,8-substituted PCDDs/PCDFs and the three non *ortho* PCBs (IUPAC numbers 77, 126, 169) was added to the tissue sample which was extracted with acetone:hexane (2:1 v/v). Water was added for phase separation followed by a further extraction with hexane. After an aliquot of the dried hexane extract was removed for total lipid determination gravimetrically, the bulk of the lipid in the remaining extract was removed by partitioning with strong sulfuric acid. The defatted sample was purified by chromatography on acid-base silica columns and separated into two fractions on magnesium silicate. The first fraction (1% dichloromethane in hexane) containing the major PCBs was concentrated to a small volume over a period of 4-6 days at room temperature, the internal standard (1,2,3,4,6,7,9-heptachlorodibenzodioxin) added, and the individual PCB congeners identified and quantified on a non-polar DB-5 column. Twenty major congeners were estimated with a limit of detection for fat samples for most congeners of about 10 μg/kg (ppb). Mono *ortho* congeners 123 and 167, minor components overall, could not be quantified due to overlap on this GC column with PCBs 118 and 128, respectively. The second Florisil fraction (dichloromethane elution) containing the PCDDs/PCDFs/non *ortho* PCBs was purified further by adsorption on fine carbon dispersed on silica gel followed by desorption with toluene. Identification and measurement of analytes in this extract have been reported[10]. Detection limits on a lipid basis of the individual congeners by MS was about 1-3 ng/kg (ppt) for fat and about 10 ng/kg for liver.

3. Results and Discussion

Table 1 summarizes the dioxin-like compounds, expressed as TEQ on a lipid basis, for the paired fat and liver tissues from the thirteen deceased individuals. This group consists of 8 women and 5 men with a mean age of 68 yr (range 45-83 yr). The overall TEQ for fat is about 750 ng/kg on a lipid basis, a value three times higher than those of Inuit residing in northern Quebec[6] and some twenty fold greater than urban dwellers from southern Quebec[11]. Using the scheme of division of dioxin-like activity as described, the proportion of the total TEQ in both tissues is in the following ascending order: 1) PCDD and PCDF, 2) non ortho PCBs, 3) mono and di *ortho* PCBs. Such a pattern with the PCB component dominating over the PCDD/PCDF appears to be typical of those people who consume large quantities of marine wildlife which in turn usually contains significantly more PCBs than PCDDs/PCDFs.

Concentrations of TEQ in adipose and liver tissues on a lipid basis are relatively similar for the PCBs. However, concentrations of the PCDDs and, more so, of the PCDFs are higher in the liver tissues than in fat. Whether this partitioning of body burden corresponds to that predicted using the toxicokinetic model developed by Carrier et al.[12] is presently being evaluated. Cytochrome P-450 protein determinations in liver samples are also underway to investigate the

relationship between body burden of dioxin-like compounds and cytochrome P-450 enzyme induction.

4. Conclusion

Exposure of Greenland Inuits to PCDDs/PCDFs/PCBs as measured in fat and liver tissue and expressed in TEQ on a lipid basis appears to be 3 times greater than Inuit from northern Quebec and markedly higher than urban dwellers from populated areas. These Inuit are one of the most exposed non occupational groups to these compounds and consist of a population with a notable chance for detection of health effects.

5. References

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Table 1. Dioxin-like compounds expressed as TEQ in omental fat and liver tissues of thirteen Greenland Inuits; values in ng/kg (ppt) on lipid basis.

Analyte	PCDDs		PCDFs		non ortho PCBs		mono & di ortho PCBs		Total TEQ	
Number of congeners	7		10		3				30	
Tissue / Individual	Fat	Liver	Fat	Liver	Fat	Liver	Fat	Liver	Fat	Liver
1	32.8	52.3	47.1	284.7	87.3	153.2	123.5	220.9	290.7	711.1
2	103.3	123.9	141.9	541.3	127.6	124.7	476.7	328.8	849.5	1118.7
3	47.6	61.6	39.7	207.7	71.0	108.6	266.6	197.2	424.9	575.1
4	67.4	70.3	102.4	98.4	133.0	99.7	381.5	394.1	684.3	662.5
5	120.5	87.3	177.1	127.4	644.3	328.3	647.3	498.1	1589.2	1041.1
6	66.3	57.6	91.2	93.0	223.2	148.9	647.4	443.3	1028.1	742.8
7	38.4	38.1	54.4	82.8	138.9	122.9	372.1	328.0	603.8	571.8
8	77.0	79.8	118.0	204.1	242.1	210.8	319.0	266.5	756.1	761.2
9	61.0	69.8	94.7	135.4	179.1	142.7	365.3	367.0	700.1	714.9
10	54.4	753.8	76.5	1090.9	112.7	1574.6	257.9	287.8	501.5	3707.1
11	40.6	51.7	51.8	230.6	209.3	292.0	399.6	362.5	701.3	936.8
12	28.9	24.6	44.2	56.4	96.4	80.4	390.0	331.4	559.5	492.8
13	87.6	92.1	109.8	180.2	493.7	381.4	371.7	369.5	1062.8	1023.2
Mean	63.5	120.2	88.4	256.4	212.2	289.9	386.0	338.1	750.2	1004.6
Geo mean	58.2	75.6	79.6	180.7	170.4	193.3	358.7	328.1	688.7	853.6