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BRIEFING:

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Twenty two samples of fish oils were analysed for their content of selected organochlorine pesticides and PCBs. Seventeen of the samples were obtained over the counter and were marketed as dietary supplements. Four were samples described as for industrial use while one was marketed for veterinary purposes./ Fish oils have wide application as dietary supplements and in the baking industry in Europe. They account for around 2% of the total world fat and food oil production and are used also in the manufacture of commodities such as margarine and ice cream. In addition they have been investigated as therapeutic agents in the treatment and prevention of cardiovascular disease, bowel cancer, psoriasis and hypertension.

Twenty one of the samples were found to contain detectable residues of organochlorine pesticides and PCBs. The levels of contamination varied widely and there appeared to be no difference between the contamianat levels present in food grade oil and industrial/veterinary grades. The most contaminated was a preparation of salmon oil marketed as pharmaceutical grade. This contained 140 microgrammes per litre of total DDT and 1132 microgrammes per litre of PCBs. Several samples of cod liver oil were also highly contaminated with up to 1055 microgrammes per litre of PCBs. Only one sample contained no detectable residues of organochlorines, and five contained comparatively low levels.

Comparison of the DDT & PCB results with those published in other studies showed that levels of total DDT appear to have generally fallen since 1986. PCB results were lower than levels previously reported from the North and Baltic Sea. Levels in cod-liver oil, however, were comparable with previous samples of this oil obtained in Icelandic waters. Currently, some manufacturers of dietary supplements are known to source their oil from the North Atlantic and Arctic regions and the current results suggest, therefore, that levels of PCB contamination are remaining stable in these regions. Changes in sourcing of the oils, the types of oils marketed and changes in processing methods are probably responsible for any changes observed between these samples and those reported earlier in the literature.

The results were used to calculate the dietary contribution of PCBs from fish oils when consumed at the manufacturers recommended levels. For the worst contaminated oil, calculated intake was 12,800% of the UK estimated daily intake published by the World Health Organisation and around 6·4% of the US Food and Drug

Administration Consumption Guideline Maximum. When used for therapeutic purposes, prescribed doses are much higher, and in the case of treatment using the most contaminated oil, the intake would reach 80% of the FDA Guidline Maximum. This is before contributions from sources such as meat and dairy products are included.

Currently, in the UK there are no set limits for PCBs in food. The only provision is the general one under the Food Safety Act (1990) that food should be safe for consumption. Similarly there appear to be no set limits for pesticides residues in fish oil in the UK. Sampling and analysis by Government Agencies has been sporadic, with the last published results dating to analyses carried out in 1991. On an international basis the Codex Alimentarius, published by the World Health Organisation carries no provision for residue levels of pesticides in fish oil.

It is clear that under certain circumstances fish oils could contribute significantly to the dietary intake of PCBs and organochlorine pesticides and there is clearly a need for an exhaustive evaluation of this. In addition, since the substances analysed are only a limited set of the toxic, persistent and bioaccumulative compounds which could be present in fish oil, there is a need to extend the chemicals analysed to include the chlorinated dioxins, toxaphene, chlordane and heptachlor to assess the full scope of the problem.

ABSTRACT

Oil rendered from whole fish and from fish offal are an important primary dietary item in many areas of the world. In addition, fish oils are used extensively in the food industry as raw materials and ingredients. An extensive, specialist market also exists for fish oils in dietary supplements. Traditionally, benefits have been conferred by their high vitamin D content in the prevention and cure of rickets in children. Therapeutic benefits in the treatment of cardiovascular, arthritic and dermatological disease have also been identified. Fish oils, however, are also susceptible to contamination with lipophilic organic chemicals. Many organochlorine chemicals are now ubiquitous contaminants of marine ecosystems. This paper reports analytical results for a selection of fish oil samples, compares the values with historical samples and discusses their potential contribution to dietary intakes of organochlorine chemicals.

Keywords: Fish-oils, PCBs, pesticides, organochlorines, diet

INTRODUCTION

Fish oil accounts for around 2% of the total world production of oils and fats, currently estimated at 80 million tonnes. (Bimbo & Crowther 1991). The largest use of fish oil is in the partially hydrogenated form in the baking industry, principally in Europe. Ockerman (1992), nonetheless, lists a variety of other applications of fish oils. These include pharmaceutical grade nutritional supplements, as raw material and ingredients for foodstuffs intended for human consumption (margarine and ice cream), in cosmetics and in detergent and soap products. In addition, industrial usages include manufacture of animal feedstuffs, the industrial processing of leather and metals, in paints, oil fabrics, printing inks, rubber, lubricants and insecticides.

Fish oils are more complex than oils derived from terrestrial animals or plants, containing a high proportion (75%) of long-chain unsaturated fatty acids. Despite this they are amenable to processing and like other edible fats and oils can be hydrolysed, saponified, hydrogenated, oxidised and sulphated. The extraction procedures differ in detail according to the fish tissues requiring processing but may *inter alia* consist of direct steaming, cold extraction, flotation or solvent extraction (Ockerman 1992). In the case of cod liver oil 3 basic grades are produced. Concentrates can be also be produced by extraction of the oils from the comminuted bodies of whole fish (Heimann 1982) and these are used for a range of purposes similar to fish liver oils.

Refining of fish oils involves the removal of free fatty acids (to facilitate onward processing operations) and stearine (to prevent clouding at low temperature) together with pigments and odours. Such refining involves various thermal processes. These processes include hot water washes and vacuum steam distillation. Solvent extraction may also be used to remove free fatty acids and natural or activated clays and carbons used to remove pigments (Ockerman 1992). In the case of cod-liver oil, 3 basic grades are produced: Pharmaceutical grade (No: 1) oil is extracted by thermal rupture of the liver cells at 82-87°C and is high in vitamin A. Number 2 oil is extracted by further steaming and pressing and is reddish in colour due to oxidation while the final grade, known as cod oil, is dark coloured and extracted from partially decomposed livers. This latter grade is used exclusively for industrial purposes.

The semi-volatile nature of the polychlorinated biphenyls (PCBs) and organochlorine pesticides (see e.g. Ballschmiter *et al.* 1989; Hansen 1994) means that these compounds are likely to be at least partially removed by such stages in the process. Nonetheless, residues of organochlorine pesticides and PCBs have been found in the few documented analyses of fish oils. Kannan *et al.* (1992) analysed pharmaceutical grade cod liver oil from the Southern Baltic to examine temporal trends in concentrations of persistent organochlorines. Samples were taken between 1981 and 1989. This showed that while DDT and DDE levels had generally declined, the PCB levels had remained steady in recent times. Later work (Falandysz *et al.* 1994a) suggested that PCBs are reaching remote areas such as the South Iceland Shelf through atmospheric cycling and are appearing, together with organochlorine pesticides, in fish oil products in appreciable quantities. On the basis of these analyses, Falandysz (1994) suggests that in the Baltic region PCBs are cycling in a steady state. By contrast, the long range transport of these substances to remote waters implies that global equilibrium has not yet been reached (Ballschmiter *et al.* 1989) although peak inputs occurred in the 1960's (Sanders *et al.* 1994).

Organochlorine contaminants present in fish-oils available from retail outlets have not apparently been subjected to independent scrutiny. This study therefore reports the results of analysis of samples of oil obtained through retail outlets as sold for dietary purposes and purchased over a period between late 1994 and early 1995 together with analyses of oils intended for industrial applications. These values are compared with those obtained from fish-oils reported in other studies. The potential contribution of fish oils to dietary intakes of organochlorines under normal conditions and under prescribed medical regimes is discussed.

MATERIALS AND METHODS

Organochlorine pesticide and PCB residues were determined according to the following method at an external laboratory. 5ml of HPLC grade hexane (Rathburn Chemicals) were added to 1ml of fish oil sample contained in a precleaned glass universal and shaken to effect complete dissolution. 2ml of concentrated Aristar grade sulphuric acid was added to the vessel, mixed thoroughly to

ensure destruction of the lipid content and the mixture allowed to stand until the hexane and acid layers separated. This extract was then transferred onto a column comprised of 0.8g active, neutral, Brockmann grade 1 aluminium oxide (BDH, Poole). The aluminium oxide was prepared for use by heating to 700°C for 4h and subsequently deactivated by tumbling with 5% distilled water by weight for 1h. The column was eluted with hexane until 5ml of eluate had been collected and 1ml of this was transferred to a glass vial. To this an internal standard of 2,6-dichlorobenzonitrile was added to give a concentration of 0.04 $\mu g\ ml^{-1}$ and the extract analysed.

Chromatographic analysis was carried out using a Varian 3400 gas chromatograph (GC) equipped with an 8035 autosampler and electron capture detector (ECD). 2μl of sample was injected in splitless mode through a split/splitless injection port held at a temperature of 200°C. The GC was fitted with two 30m DB210 columns (J&W Scientific) connected in series using a 4m deactivated retention gap, of 0.253mm internal diameter and 0.25μm phase thickness. The carrier gas was hydrogen (Distillers MG, Grade 5.0) at a velocity of 51cm s^{-1}. The temperature programme of three levels comprised a first level of 185°C ramped at 2.5°C min^{-1} and held for 20 minutes. This was then increased to 210°C at 5°C min^{-1} and held for twelve minutes before reaching the final temperature of 240°C at a rate of 40°C min^{-1} to give a total run time of 87.75 min.

Identification and quantification of analytes was by comparison with prepared calibration standards. This operation was performed using the EZCHROM Version 5.2 chromatography data system. Recoveries of analytes from the alumina columns were checked and exceeded 96% in every case. Recoveries of analytes from samples spiked with 50μl of a prepared standard solution and subjected to the full analytical procedure were also checked and these recoveries are shown in Table 1. A multilevel calibration was carried out to determine the linearity of the ECD over a concentration range of 20-100ng ml^{-1} for each component. The ECD response factor was determined prior to analysis of each batch of samples using a single working standard containing 80ng ml^{-1} of each analyte.

In order to facilitate comparisons between the data expressed here in $\mu g/l$ with previously published data expressed in $\mu g/kg$, densities of oils were determined gravimetrically. These were found to lie between 0.911 and 0.937 g ml^{-1}. In addition, to allow better evaluation of the influence of oil type upon contaminant levels, the individual pesticide and congener specific data were subjected to principal components analysis using the MINITAB software package. The resulting principal component plots are presented as FIGURES 2 & 3.

RESULTS

With the exception of sample 16, a marine lipid concentrate, all samples of fish oils examined contained detectable residues of organochlorine contaminants. Results for chlorinated pesticides

and total PCBs are shown in TABLE 2 while congener specific analyses are reported in TABLE 3. Mean values, standard errors and ranges of the specific analytes are shown in TABLE 4. The values obtained for cod liver oil are generally lower by an order of magnitude than those reported by Falandysz *et al.* (1994) for the Baltic. The oil (Sample 18) derived from the Iceland Shelf is described as a fish oil and is not therefore directly comparable with the previously published values. Values are also very much lower than those reported by the same authors for cod liver oils from the Southern Baltic (see: TABLE 4.) The highest overall contaminant concentrations were found in a salmon oil preparation (Sample 9). This may not be representative of the true content of salmon oil since this oil is often extracted using pilchard or Greyfish oil (Ockerman 1992) followed by simple centrifugation to separate the oil mixture. Equally, it may reflect the use of farmed salmon reared in coastal waters and fed on synthetic foods.

After the salmon oil, cod liver oil samples were the most highly contaminated (Samples 8, 13, 15, 20) although this group of 8 contained the lowest contaminated oil after Sample 16 (Sample 3). Sample 20 is included on the grounds that it is probably a No: 2 cod liver oil. Similarly, elevated levels were found in the fish oil concentrate (Sample 17). Products described as fish oils (Samples 1,2,3,4 & 5) including the sandeel oil (Sample 21) had intermediate to low levels of contamination together with the halibut liver oil (Sample 12). According to the list of ingredients the packaging for Sample 12, this oil was a mixture of unknown proportions of halibut liver oil and soya oil. The levels of contaminants found therefore cannot be regarded as representative of halibut liver oil alone. No organochlorine contaminants were detected in Sample 16 and comparatively low concentrations were found in Sample 11, another marine lipid concentrate, comparable with Sample 3.

Quantitatively, the dominant contaminants are the PCBs, followed by DDT and its metabolites DDE and TDE. TDE is also known as DDD. Hexachlorobenzene (HCB) and Hexachlorcyclohexane (HCH) isomers are lesser components of the overall contaminant content and are absent from many samples. Dieldrin residues are not reported since recovery tests showed that this contaminant did not survive the acid clean-up process. The levels of contaminants present in the industrial and veterinary grade oils do not appear markedly different from concentrations present in those oils designated as pharmaceutical grade.

The same general trend is also true of PCBs analysed on a congener specific basis (TABLE 3). The congener specific values obtained are broadly comparable to values obtained from a fish oil derived from a batch of North Sea fish for intercalibration purposes (R1). Again, concentrations are generally an order of magnitude or more lower than recorded for cod liver oil in 1983 from the North Sea (R4). This oil was supplied from the same manufacturer as Samples 1 & 2. An order of magnitude difference generally exists between values reported for a 1989 Canadian Reference Material and the present samples. Organochlorine concentrations in the more contaminated cod liver oils, nonetheless, are of a comparable order of magnitude to those determined for a 1984 sample obtained

from Iceland and for which a ΣPCB value of 1.9ppm is given (Falandysz 1994).

There are some further differences: CBs 28 and 52 were not detected in any of the samples analysed in this study although this may be a function of relatively high detection limits determined for these congeners. While CBs 138 and 153 were generally the dominant congeners in the cod liver oil samples, the relative proportions do not appear to be as high as in samples R1-R5. Overall, CB 118 appears to comprises a higher proportion of the congener content than for the various reference samples shown for comparison in TABLE 3. Moreover, the relative proportions of the congeners are highly variable between the samples as shown in FIGURE 1. Principal components analysis (FIGURE 3) suggested that samples with high CB 101 and CB 149 values have relatively lower values of CB 128 and 153. This analysis, however, is probably confounded by the inexplicably high value for CB 101 in Sample 9. There are a number of possible reasons for the above differences which are discussed in further detail below.

DISCUSSION

Results from the analysis of fish oils indicate that none exceed the 2.0 ppm regulatory limits specified for foodstuffs by a number of authorities including the US Food and Drug Administration (see: Simmonds *et al.* 1994) although in some cases the 1.0 ppm limit specified in Switzerland is exceeded. In the UK there are no specific legal requirements relating to the PCB content of food, including fish oils, except for the general provision enshrined in the Food Safety Act (1990) that commodities should be safe for human consumption. Further, there appear to be no specific limits set for pesticide residues for fish oils in the UK. Similarly, the Codex Alimentarius published by the United Nations World Health Organisation appears to contain no recommendations for limits on organochlorine pesticides or PCBs in fish oils although oils derived from fish and marine mammals are recognised as a commodity group (FAO 1993).

Comparison of the values obtained is made difficult by the fact that the oils are undoubtedly of different origins in terms of both subject species and geographical location. Principal components analysis (FIGURES 2 & 3) shows no clear groupings of the oils by geographical origin or oil type with respect to the contaminant levels found. Both these factors are likely to influence concentrations of contaminants in fish oils. This also holds true for comparisons with historical data. Further, most published data relates specifically to cod liver oils rather than other fish oils or concentrates which are extracted from the whole bodies and livers of a variety of species.

Overall, the levels of organochlorine contaminants present in the fish oils analysed in this study appear somewhat lower than those reported by Falandysz *et al.* (1994a) for cod liver oils derived from a variety of areas between 1981 and 1989 and usually sampled annually from bulk storage tanks. Results for Sample 1 are much lower than the values given in row R4 of Table 3 which shows the

analytical results from a sample of oil obtained in 1983 from the same supplier. Levels of PCBs determined on a congener specific basis are in general an order of magnitude lower in the 1994 sample than in that obtained in 1983 although of the same order of magnitude as a 1984 sample obtained in Iceland. Concentrations of ΣDDT are substantially lower than previously reported figures for the Shelf of Iceland (TABLE 4) samples taken between 1984 and 1987. Similar findings were made for organochlorine pesticides between samples analysed in 1985 and 1991 (MAFF 1992). The levels of DDT found in the 1991 survey are comparable to levels found in this study although HCB levels appear somewhat lower. A total of 22 of the 29 samples analysed by MAFF (1992) contained organochlorine pesticide residues but PCBs were not analysed.

Although controls on both PCBs and the organochlorine pesticides may be partially responsible, it is probable that changes in the sourcing of oil and changes in processing methods have also contributed. For example Falandysz *et al.* (1994b) note that restrictions on use of cod-liver oil from the Baltic Sea were emplaced in Poland in 1982 although no controls exist on imported products. In the UK some suppliers have specifically stated that their oil is sourced only from North Atlantic/Arctic waters, while others exclusively use oil taken from the Norwegian Sea and Pacific Ocean sources (Seven Seas Ltd pers. comm; Lamberts Healthcare pers. comm.). Blending of oils as noted for Sample 12, can also dilute the content of contaminants present in unblended oil. Changes in sourcing of fish oils will undoubtedly be reflected in changes in contaminant levels given the differences found by Falandysz *et al.* (1994a) between samples of cod liver oil from different regions and the inter- and intra- hemispheric differences known to exist in ambient environmental organochlorine contamination (see: Tanabe 1988).

Although restrictions on the use of PCBs and many organochlorine pesticides have resulted in a decline in local environmental levels (see: IJC 1989; Sanders *et al.*1994) contaminated areas such as the Great Lakes now appear to be acting as sources of, for example, PCBs into the wider environment (Swackhamer & Eisenreich 1991). Overall, Loganathan & Kannan (1994) consider that, although restrictions on use led to rapid declines of organochlorines in freshwater biota, semi-closed and coastal waters show relatively slow clearance rates. They consider that open ocean waters are serving as a sink for persistent organochlorines. Moreover, PCBs are still in extensive use and only a small proportion have been destroyed (Tanabe 1988). DDT and τ-HCH are still widely used although restricted or banned in many countries (Voldner & Li 1995). In addition, it has been convincingly postulated (Wania & McKay 1993) that northern latitudes may be acting as a sink for semi-volatile organochlorine contaminants as part of a global equilibration process. This hypothesis is substantiated by empirical data obtained from these regions (Muir *et al.* 1992).

It is unlikely, therefore, that the observed fall in organochlorine levels particularly in oils derived from fish in northern latitudes is attributable to regulatory effort alone. If anything concentrations of PCBs could be expected to remain relatively stable. Indeed the time series of samples analysed by

Falandysz (1994) for the highly contaminated Baltic Sea have shown that levels in cod liver oil remained stable there between 1971 and 1986 despite regulation. The comparability of the more highly contaminated cod liver oils with R5 on a PCB congener specific basis may thus reflect the fact that these oils are indeed now being sourced predominantly from the North Atlantic open water fisheries.

Marketed fish oils also generally undergo some degree of refining which may also remove trace contaminants. The analytical results suggest that changes in the processing regime used by the manufacturers may also be partially responsible. A feature of the analysed oils in this study was the non detection of CBs 28 and 52 which are relatively volatile congeners. A variety of the processes used in fish oil production have the potential to remove semi-volatile organochlorine contaminants. In particular, solvent extraction procedures followed by vacuum distillation (Ockerman 1992) may be effective in this regard. Steam distillation is also employed to deodorise fish oils and this effectively removes volatile components responsible for odour. This process is also likely to remove semi-volatile organochlorine contaminants. Deodoriser distillate arising from this step could, therefore, result in a significantly contaminated waste stream. The problem of organochlorine contaminants and the need for their removal has been recognised in the industry and innovative solutions proposed for removing these substances include the use of supercritical carbon dioxide (Jakobson *et al.* 1991).

The results obtained in this study suggest that despite controls on the use of persistent organochlorine substances and the possible introduction of manufacturing refinements to remove them, appreciable quantities of these contaminants are found in pharmaceutical and industrial grade oils available on the open market. There appears to be little regulatory control and few advisory limits appear to have been published. Many of these oils are intended for dietary uses by the healthy consumer.

The properties of fish oils have also led to them being investigated and used for therapeutic purposes. It is of interest, therefore, to examine the potential contribution to dietary intake of organochlorine compounds using the PCBs as an example. In this context, it is important to note that organochlorine intakes will result from ingestion of other foods, principally other fish products together with meat and dairy products (Grove *et al.* 1992; Himberg *et al.* 1993; Theelen 1991). Moreover, fish oil can act as a source of organochlorines in addition to those measured in this study such as the chlorinated dioxins (Theelen 1991) and toxaphene (de Boer & Wester 1993). These are also of concern and have been detected at significant concentration in fish livers and in the oils derived from them.

TABLE 5 shows the manufacturer's recommended daily consumption of fish oil as indicated on the relevant packaging together with an intake figure for total PCBs and DDT calculated from the results obtained by analysis of the specific packaged products. TABLE 6 shows estimated daily intakes of PCBS from dietary sources taken from Moy *et al.* (1992). In turn these are taken from the estimated

intake of PCB figures from dietary sources as given in the UNEP/FAO/WHO Food Contamination and Assessment Programme. These range from 0.0005 μg kg body weight^{-1} day^{-1} for the UK to 0.9 μg kg body weight^{-1} day^{-1} for New Zealand. This high value is attributed to high consumption of dairy products. Fish consumption in Finland and the Netherlands is thought to be responsible for the relatively high intakes in these countries. Nonetheless, the figures themselves are the subject of considerable uncertainty. Many were derived before modern analytical techniques were fully evolved. In addition the methodologies employed to construct the estimates may result in wide differences. As an example, the two estimates derived for Finland used different methods and differ by an order of magnitude (Table 6). Similarly, the 1981 value for the UK of 0.0005 contrasts with the value of between less than 0.14 and 0.57 μg kg body weight^{-1} day^{-1} based on a 70 kg adult (MAFF 1983).

On the basis of the calculated results presented in TABLE 5, consumption of fish oil at manufacturers recommended levels can result in daily intakes of PCBs between 0.009 and 4.5 μg day^{-1} or between 0.12x10^{-3} and 0.064 μg kg body weight^{-1} day. This excludes Samples 3, 5 and 16 in which PCBs were not detected. At the highest level this exceeds many of the estimates presented in TABLE 6. At intermediate levels of recommended intake or with excedence of recommended consumption levels, fish oils could account for a significant proportion of estimated PCB intakes. The wide variation in contaminant levels in the oils together with the varied recommended intakes means that a meaningful comparison using the mean value calculated is not possible. On the basis that Sample 9 of salmon oil represents a worst case at an intake of 0.064μg kg body weight^{-1} day^{-1}, then this represents a potential contribution of between 7 and 12800% of the estimated daily intakes given in TABLE 6. This also comprises 6·4% of the US FDA Maximum Consumption Guideline of 1μg kg body weight^{-1} day^{-1} (Moy *et al.* 1993). This is excluding the possible contribution from other foods.

Fish oils have also been identified as of potential therapeutic value due to their high content of omega-3 fatty acids and their contents of vitamin A, D & E. The treatment of rickets and promotion of wound healing using fish oil is well known (Falandysz *et al.* 1994b) but fish oils have also been investigated for the treatment and prevention of *inter alia* cardiovascular disease (Lee *et al.* 1985; Kestin *et al.* 1990; Turini *et al.* 1994; Mori *et al.* 1994) rheumatoid arthritis (Kremer *et al.* 1987), hypertension (Knapp & Fitzgerald 1989; Sacks *et al.* 1993; Passfall *et al.* 1993), psoriasis (Grimminger *et al.* 1993) and colon cancer (Bartram *et al.* 1993). In these applications, prescribed dietary intakes may be substantially above the levels associated with normal dietary supplementation. TABLE 7 gives prescribed intakes from a variety of clinical studies.

From these figures it is obvious that the quantities of fish oil used in therapeutic applications may contribute substantially to intake of organochlorine contaminants even when the oils are only moderately contaminated. Excluding the emulsion supplied intravenously, daily volumes prescribed range between 6 & 50ml of

oil. At the highest contamination level of 1132 $\mu g\ l^{-1}$ this translates to a total intake of up to 56.6 $\mu g\ day^{-1}$ or about 80% of the US FDA Consumption Guideline. Again, this does not consider intakes from other dietary sources.

In conclusion, the levels of organochlorine contaminants present in fish oils from various sources could constitute a significant contribution to daily intakes of organochlorine contaminants, particularly the PCBs used an example in this case. This is without consideration of other potential dietary sources of PCBs and other organochlorines. Those consuming large quantities of fish oils for therapeutic purposes may have substantially higher intakes. It follows that contamination of fish oils requires careful monitoring if intended for use as a dietary supplement, therapeutic agent or in the preparation and manufacture of other foodstuffs.

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ANALYTE	%RECOVERY
HCB	110.4
α-HCH	86.0
γ-HCH	73.5
p,p'-DDE	124.5
p,p'-TDE (DDD)	85.0
p,p'-DDT	91.9
CB-8	97.1
CB-18	104.6
CB-28	107.7
CB-31	110.1
CB-52	93.2
CB-77	90.1
CB-101	90.2
CB-118	77.1
CB-126	91.1
CB-128	109.2
CB-138	93.5
CB-149	93.5
CB-153	101.1
CB-169	88.9
CB-170	91.1
CB-180	104.0
Aroclor 1254	89.2

TABLE 1: Percentage recovery of analytes from fish oil samples spiked with a recovery standard of mixed organochlorines. Chlorobiphenyls are numbered according to Ballschmiter & Zell (1980).

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SAMPLE	ORIGIN	GRADE	TYPE	HCB	a-HCH	g-HCH	p,p'-DDE	p,p'-TDE	p,p'-DDT	ΣDDT	ΣPCBs
1	Norway	P	FO	-	-	-	45	29	74	148	570
2	Norway	P	FO	-	-	-	30	19	14	63	440
3	Norway	P	CL	-	-	-	2	-	-	2	-
4	Japan	P	FO	2	6	-	62	-	33	95	313
5	Japan	P	FO	-	-	-	2	-	-	2	-
6	Spain	P	CL	18	93	-	59	10	-	69	261
7	Spain	P	CL	2	-	-	22	6	12	40	212
8	UK	P	CL	21	9	3	56	44	19	119	990
9	UK	P	SO	46	8	9	87	53	-	140	1132
10	UK	P	CL	-	-	-	28	27	7	62	428
11	UK	P	MLC	-	-	-	4	-	-	4	10
12	UK	P	HL	-	-	-	3	-	-	3	37
13	UK	P	CL	10	12	3	60	48	29	137	1055
14	UK	P	CL	-	7	-	6	-	-	6	14
15	UK	P	CL	10	12	-	60	47	31	138	1050
16	UK	P	MLC	-	-	-	-	-	-	-	-
17	UK	P	CFO	-	-	-	35	23	-	58	915
18	Iceland	U	FO	14	36	3	11	-	7	18	366
19	Germany	U	CFO	21	33	20	56	51	30	137	939
20	Germany	U	RF	24	18	-	33	37	29	99	1106
21	Germany	U	SE	8	14	11	14	23	-	37	463
22	UK	V	CL	10	26	-	9	-	-	9	183
DETECTION LIMIT				3	3	2	1	4	5	N/A	10

TABLE 2: Concentrations of organochlorine contaminants present in fish-oils obtained from various sources. P: Indicates pharmaceutical grade oil; U: Indicates oil for use in industrial applications V:denotes a veterinary grade oil. Concentrations are given in micrograms per litre. (-): Indicates analyte not detected. N/A: denotes detection limit value not applicable, since sum value for ΣDDT was obtained by addition of individual values for DDT, DDE & TDE, each with specified detection limits. Abbreviations: FO: Fish oil; CL: Cod liver oil; CFO: Crude Fish Oil; SE: Sandeel; RF: Redfish Oil (Probably No: 2 cod liver oil); MLC: Marine Lipid Concentrate; HL: Halibut liver oil; CFO: Fish oil concentrate; SO: Salmon oil. Descriptions are those given by the manufacturer/supplier.

SAMPLE	ORIGIN	GRADE	TYPE	PCB CONGENER										
				28	52	101	118	128	138	149	153	169	170	180
1	Norway	P	FO	-	-	15	24	9	76	16	52	-	6	22
2	Norway	P	FO	-	-	10	14	6	49	-	37	-	10	12
3	Norway	P	CL	-	-	-	-	-	6	-	-	-	-	-
4	Japan	P	FO	-	-	-	-	-	24	10	24	14	-	21
5	Japan	P	FO	-	-	-	-	-	-	-	-	-	-	-
6	Spain	P	CL	-	-	-	5	-	29	-	32	-	3	7
7	Spain	P	CL	-	-	-	-	-	27	-	24	-	14	11
8	UK	P	CL	-	-	29	119	11	110	18	70	-	12	13
9	UK	P	SO	-	-	156	60	-	61	51	-	-	11	44
10	UK	P	CL	-	-	-	36	17	43	-	57	-	10	31
11	UK	P	MLC	-	-	-	9	-	-	-	-	-	-	-
12	UK	P	HL	-	-	-	-	-	12	-	-	-	-	-
13	UK	P	CL	-	-	26	117	10	122	-	79	-	13	29
14	UK	P	CL	-	-	-	-	-	-	-	-	-	-	6
15	UK	P	CL	-	-	22	116	8	60	18	75	-	14	17
16	UK	P	MLC	-	-	-	-	-	-	-	-	-	-	-
17	UK	P	CFO	-	-	19	30	-	61	52	70	-	11	13
18	Iceland	U	FO	-	-	-	36	-	16	-	-	-	13	-
19	Germany	U	CFO	-	-	51	76	-	65	44	80	-	21	14
20	Germany	U	RF	-	-	18	79	-	32	26	-	-	31	11
21	Germany	U	SE	-	-	21	30	-	44	41	56	-	-	12
22	UK	V	CL	-	-	-	28	-	12	-	-	-	9	-
DETECTION LIMITS				11	18	7	5	6	4	8	6	4	3	3
R1	N/A	N/K	CL	15	39	54	71	N/V	94	N/V	124	N/V	N/V	38
R2	N/A	N/K	CL	28	80	129	91	179	267	105	276	49	N/V	108
R3	N/A	P	CL	10	23	45	80	N/V	160	N/V	120	N/V	N/V	50
R4	Norway	P	CL	72*	96	240	380	48	520	140+	590	N/V	57	230
R5	Iceland	P	CL	-	45	88	120	33	220	64+	250	N/V	19	87

TABLE 3: Concentrations of individual chlorobiphenyl congeners determined in fish oils from various sources. All figures are given as micrograms per litre. (-): Indicates analyte not detected. ICES congeners are 28, 52, 101, 118, 138, 153, 180 (Law et al. 1989). Values in ug/kg for reference fish oil prepared from North Sea fish and used in the QUASIMEME scheme reported by Wells & De Boer (1994) are shown as R1 corrected to two significant figures. Similarly, R2 reports concentrations of CBs (ug kg^{-1}) in a Canadian reference material issued in 1989 (from: Schantz et al. 1992). Row R3 shows mean values (ug kg^{-1}) of five samples of pharmaceutical grade fish oils analysed and reported by Himberg et al. (1993). N/V denotes no value given. R4 gives values for a sample supplied in 1983 from the same source as sample 1 while R5 is a cod liver oil sample from Iceland in 1984 and analysed by Falandysz (1994). * denotes an aggregate value for CBs 28 & 31, + is an aggregate value for CBs 144 & 149. Direct comparisons of concentrations given in µg/l with those given in µg kg^{-1} should allow for an approximate 10% underestimate of concentrations expressed on a volumetric basis. Densities of fish oils determined gravimetrically showed a range between 0.911 and 0.937 g/ml.

ANALYTE	HCB	a-HCH	g-HCH	p,p'-DDE	p,p'-TDE	p,p'-DDT	ΣDDT	ΣPCBs
RANGE	ND-46	ND-93	ND-20	ND-87	ND-53	ND-74	ND-148	ND-1132
MEAN	9.1	13.1	3.0	31.1	19.8	14.2	66.0	479.0
SE	2.4	4.4	1.0	5.5	4.1	3.8	11.8	89.0
SI	73-100	42-71	5-9	340-440	-	15-120	650-950	1.9*
Mean	87	53	6	400		76	860	N/V
SB	170-370	280-400	100-160	1500-5100	-	440-2100	3100-12000	8100-16000
Mean	280	320	140	2600		1300	6300	10

TABLE 4: Values of the range, mean and standard error about the mean for organochlorine contaminants present in fish oil shown in µg/l. ND denotes analyte undetected. Mean values calculated using one half of the detection limit shown in TABLE 2 where contaminant levels were below this value. n=22 except for ΣDDT where n=21 due to omission of one value below detection limit. Lower table shows comparative values in ug kg^{-1} for fish oils sampled between 1984 and 1989 from the Shelf of Iceland (SI) and Southern Baltic (SB) (Falandysz et al. 1994). * Denotes a single value. ΣDDT values not directly comparable due to summation of different isomers in each case.

SAMPLE	RECOMMENDED DAILY INTAKE (ml)	ΣPCB DAILY INTAKE (µg)	ΣDDT DAILY INTAKE (µg)
1	5-7	2.85-3.99	0.74-1.036
2	5	2.2	0.32
3	5	N\D	0.01
4	0.25	0.08	0.02
5	0.3	N\D	0.0005
6	-	-	-
7	0.2-1.2	0.05-0.31	0.01-0.08
8	1	0.99	0.12
9	4	4.5	0.56
10	0.25	0.12	0.02
11	1-6	0.01-0.06	0.004-0.024
12	0.25	0.009	0.0007
13	0.54	0.56	0.074
14	1	0.014	0.006
15	0.54	0.57	0.074
16	2-8	N/D	N/D
17	1-4	0.92-3.66	0.058-0.232

TABLE 5: Recommended daily dietary intakes of fish oil according to product manufacturers. Total PCB and DDT intake are shown as calculated from the analytical determinations presented in TABLE 2. (-) Indicates that recommended dosage is not known. N/D denotes residues not detected in original samples.

COUNTRY	YEAR	ESTIMATED INTAKE ($\mu g\ kg\ b.w^{-1}\ day^{-1}$)	CALCULATED TOTAL ($\mu g\ day^{-1}$)
AUSTRALIA	1987	0.002	0.14
FINLAND	1984	0.21	14.7
	1993*	0.25	15.0
	1994+	0.03	2.3
GUATEMALA	1988	0.012	0.84
JAPAN	1988	0.045	3.15
NETHERLANDS	1984	0.2	14.0
NEW ZEALAND	1982	0.9	63.0
SWITZERLAND	1983	0.12	8.4
UK	1981	0.0005	0.035
USA	1988	0.001	0.07

TABLE 6: Estimated intake of PCBS in μg kg body weight $^{-1}$ day $^{-1}$ and calculated total intake of PCBs based upon data submitted as part of the UNEP/FAO/WHO Food Contamination and Assessment Programme and reported by Moy et al. (1993). Body weight figure of 70kg assumed in the calculation except for $^{(*)}$ where 60kg applies. * & $^{+}$ denote estimates derived by two different research groups in Finland (*: Himberg et al. 1993; $^{+}$: Hietaniemi & Kumpulainen 1994).

DAILY DOSE	DURATION	INVESTIGATION	REFERENCE
6g	12 weeks	Rheumatoid arthritis	Tulleken et al. (1990)
15g	14 weeks	Rheumatoid arthritis	Kremer et al. (1987)
20g	6 weeks	Rheumatoid arthritis	Sperling et al. (1987)
9g	30 days	Rheumatoid arthritis	Magaro et al. (1988)
9g	6 weeks	Hypertension	Passfall et al. (1993)
16.5g	6 weeks	Hypertension	Norris et al. (1986)
10 & 50ml	4 weeks	Hypertension	Knapp & Fitzgerald (1989)
6g	24 weeks	Hypertension	Sacks et al. (1994)
12g	20 weeks	Cardiovascular disease	Hellsten et al.(1993)
6 & 12g	12 weeks	Cardiovascular disease	Mori et al. (1994)
18g	6 weeks	Cardiovascular disease	Lee et al. (1985)
9g	4 weeks	Blood lipids	Mantzioris et al. (1994)
13.8g	42 days	Platelet aggregation	Turini et al. (1994)
11g	2x4 weeks	Rectal cell proliferation	Bartram et al. (1993)
100ml*	40 weeks	Guttate psoriasis	Grimminger et al. (1993)

TABLE 7: Reported prescribed intakes of fish oil supplements used in clinical investigations. Time indicated specifies periods when oils were administered and does not include wash-out periods or crossover treatments. * indicates 50ml lipid emulsion prepared from marine fish oil and administered by an intravenous route twice daily.

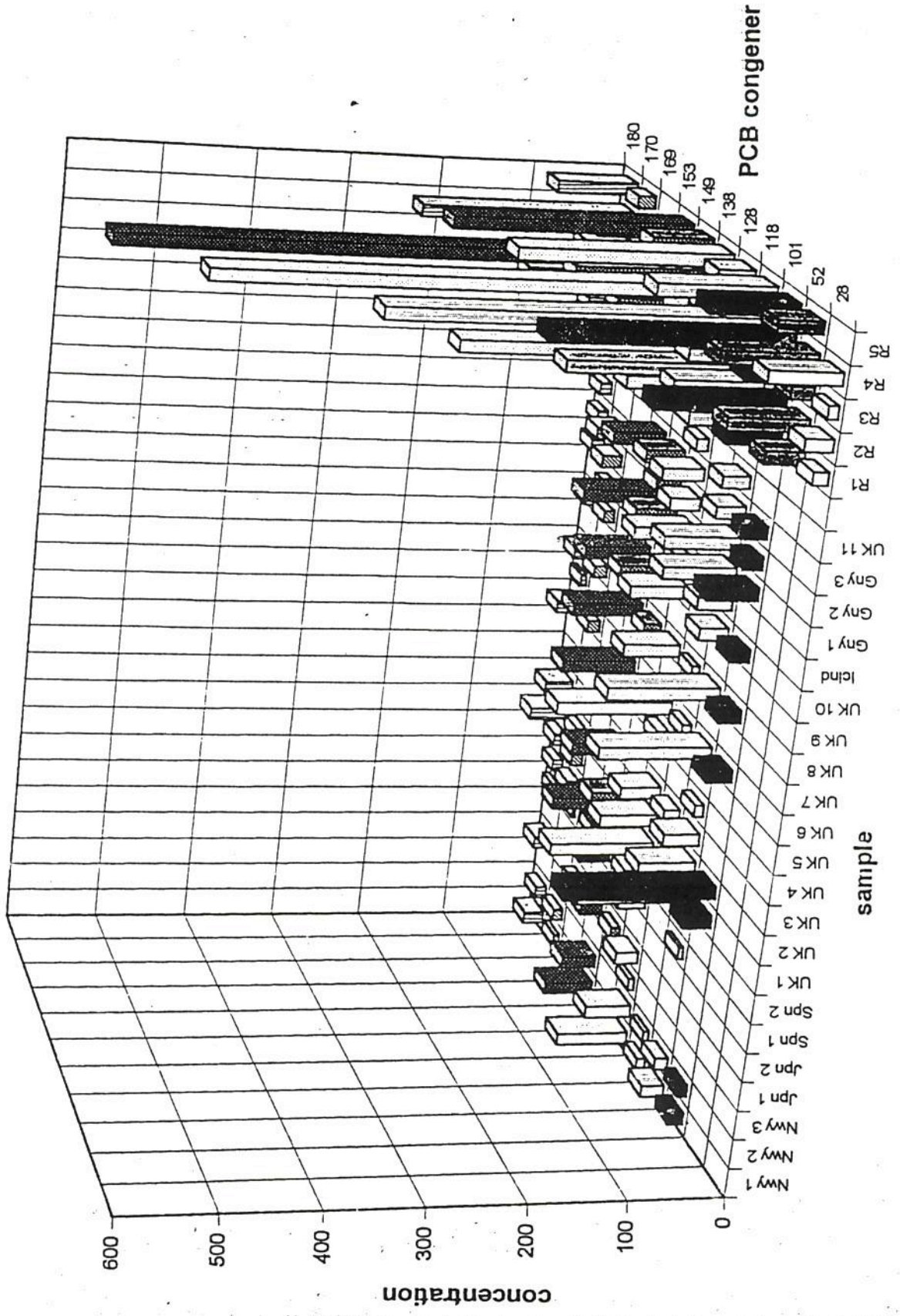
LEGENDS TO FIGURES

FIGURE 1: Three dimensional histogram showing concentrations of PCBs on a congener specific basis in μg/l of fish oil.

FIGURE 2: a) Principal Components plot for concentrations of organochlorine contaminants present in fish oils grouped by type of oil. KEY as per TABLE 2: 1: FO; 2: CL; 3: SO; 4: MLC 5: HL; 6: CFO; 7; RF: 8: SE; *: Unknown oil type. b) Principal Components Plot for concentrations of organochlorine contaminants present in fish oils grouped by country of origin KEY: 1: Norway; 2: Japan; 3: Spain; 4: UK; 5: Iceland; 6: Germany.

Figure 3: a) Principal Components Plot for concentrations of PCBs on a congener specific basis in fish oil grouped by type of oil. b) Principal Components Plot for concentrations of PCBs on a congener specific basis, grouped by country of origin. Key as for FIGURE 2.

Fig 1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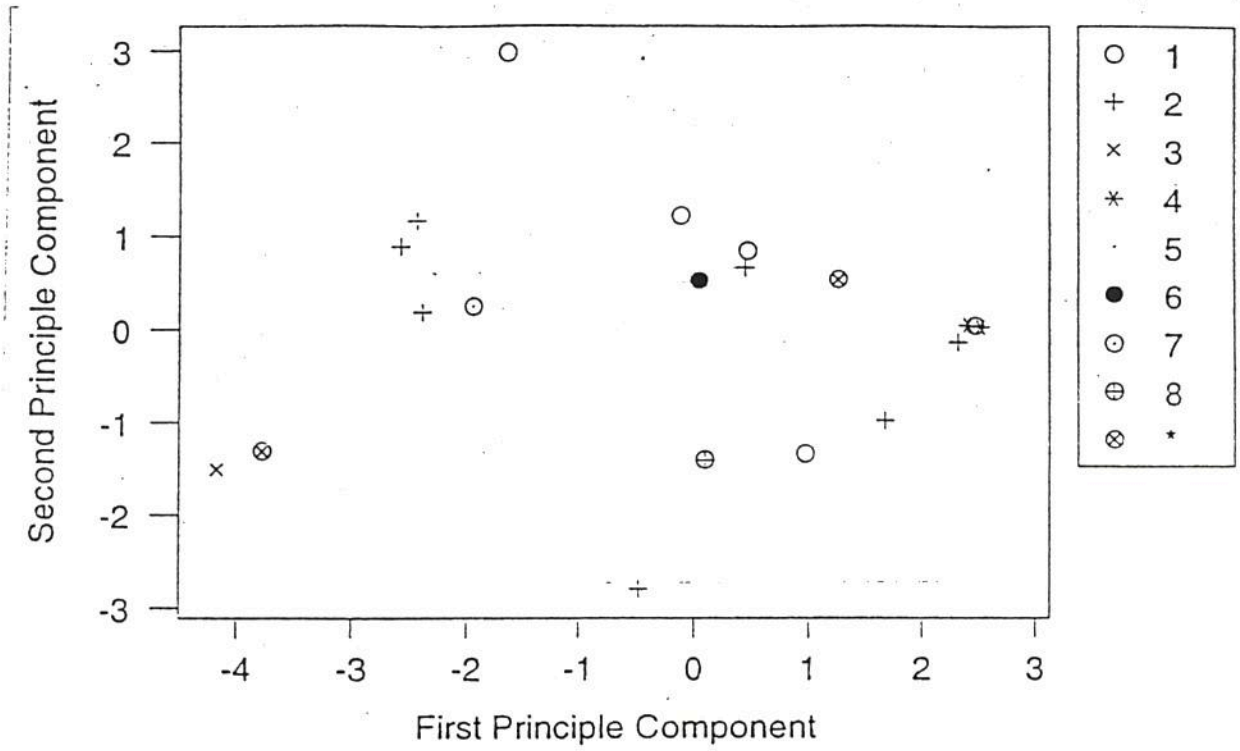
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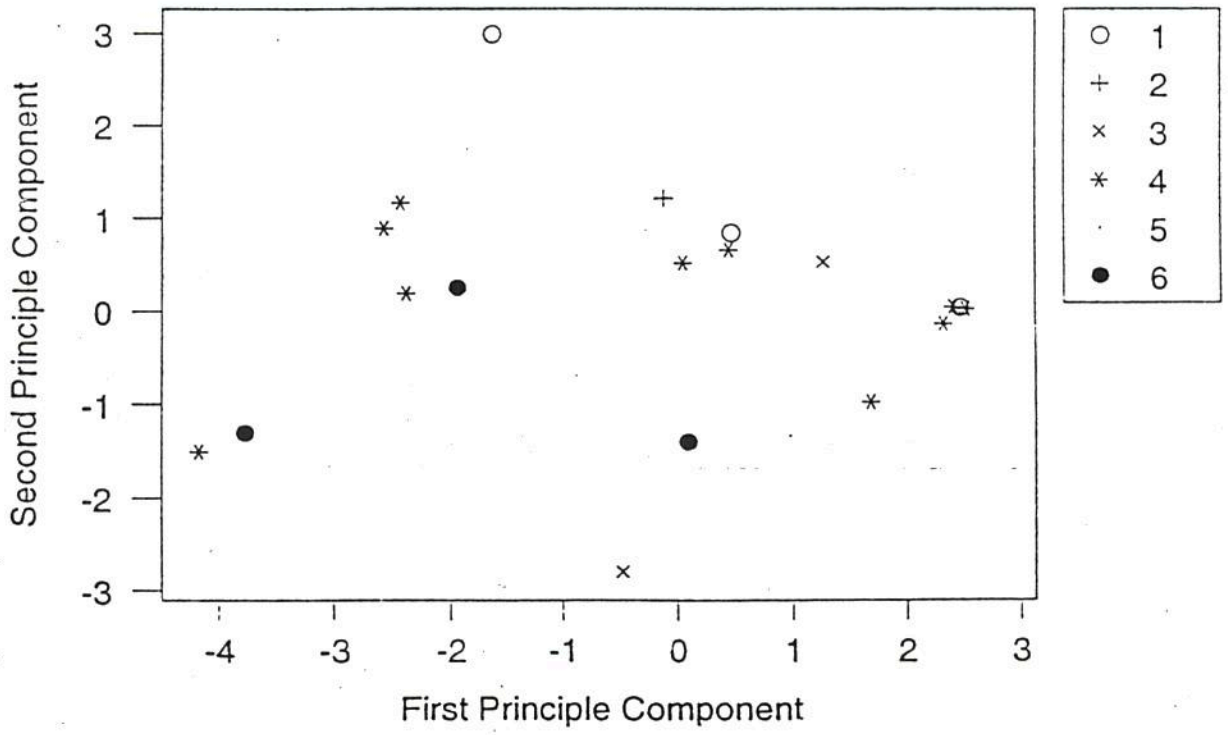
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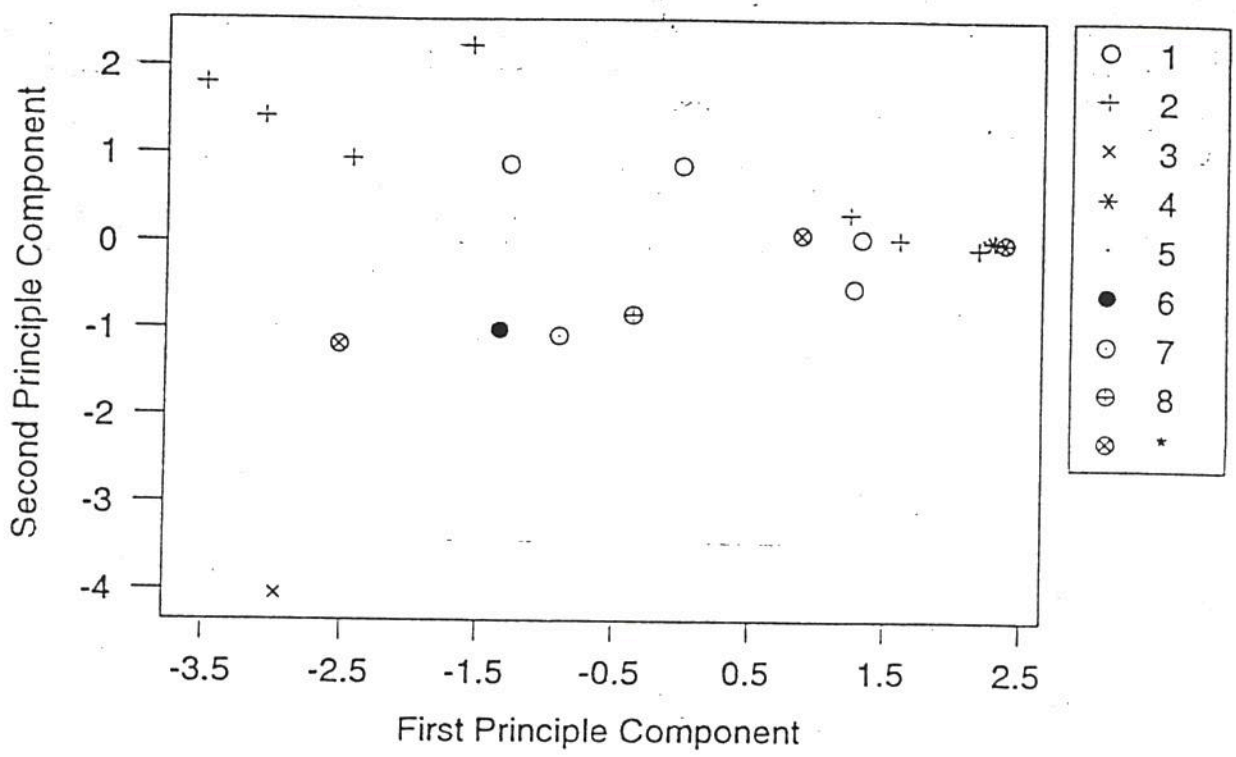
FIG 3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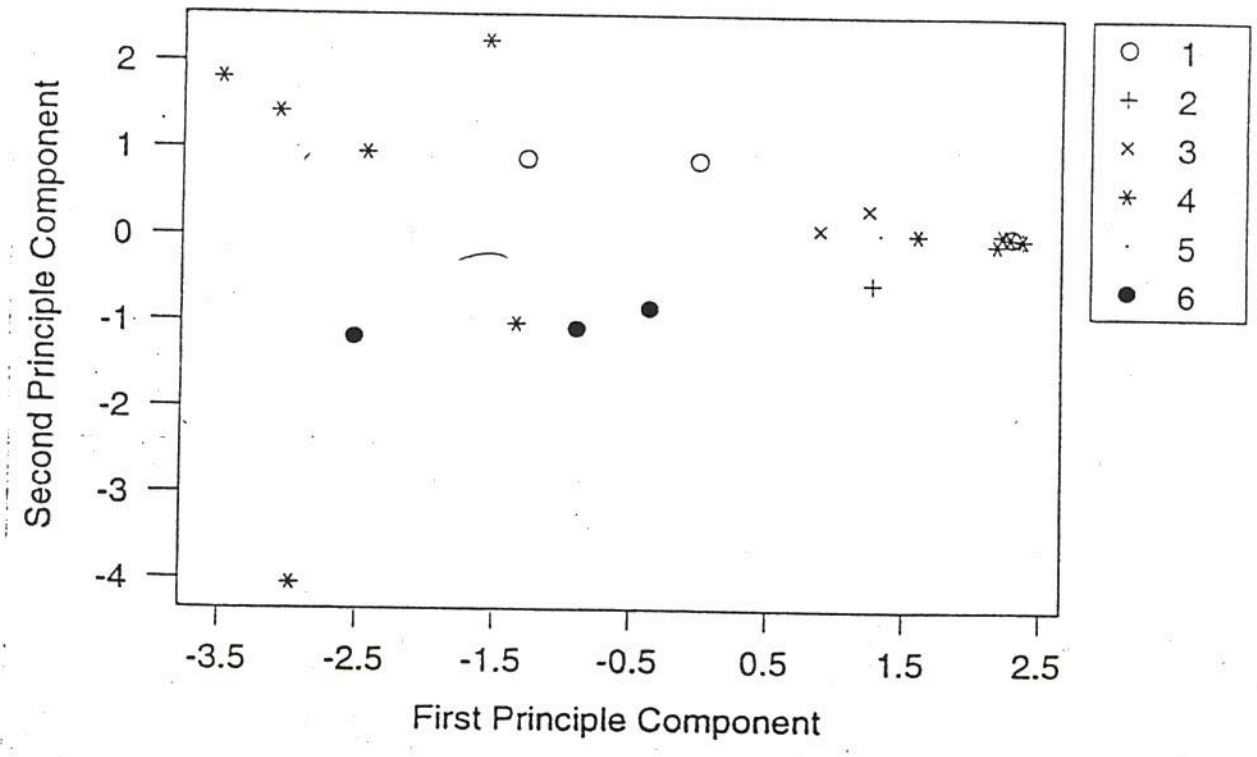
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