A critical reassessment of current human dietary exposure to PCDD's and PCDF's in the UK

Miriam Jacobs [a] and Paul Mobbs[b],
[a] Public Interest Consultants, 32 Constantine Road, London NW3 2NG
[b] Mobbs Environmental Research, 3 Grovenor Rd, Banbury, Oxon. OX16 8HN

Abstract

Current human dietary exposure to the seventeen 2,3,7,8- chlorine substituted dioxins and furans from fat containing foods in the UK were reassessed to give more reliable estimates of recent average UK dietary intakes.

Using analytical data from the UK Total Diet Survey [1] the summed toxic equivalent concentrations (TEQ) results in 11 food groups were combined with dietary fat intake data from the three main UK dietary intake survey's currently available, to give an age range of the estimated mean population dietary intake of PCDDs and PCDFs. The results were 2.93 pg TEQ/kg b.w. day for adults, or 175.5 pg/TEQ/person/day, in contrast to MAFF estimates of 69 and 88 pg/TEQ/person/day,[2] and a recently modelled value of 61 pg/TEQ/person/day, based on the US EPA food chain exposure model.[3] The levels for young children, especially breast fed infants, exceeded the Tolerable Daily Intake, ranging from 54.19 pg TEQ/kg b.w./day at 6 months of age down to 0.25 pg TEQ/kg b.w./ day at 4 1/2 years.

1. Introduction

The congener specific analyses of PCDD/F's undertaken on samples of fatty foods from the UK total diet study collected in 1982 and 1992, have been reported by The Ministry for Agriculture, Fisheries and Food (MAFF) Food Contaminants Division, UK.[2] The TDS aims to represent the relative importance of retail foods in the average UK diet using the economic consumption data from the yearly National Food Survey (NFS) together with trade statistics. Therefore the proportions of different foods in the composite food samples vary according to the year of collection and the year of the NFS. MAFF estimated the dietary intakes of PCDD/F's for average and high level percentile (97.5th) adult UK consumers by two methods; 1. combining data from the analysis of TDS samples with food consumption data from the NFS [4,5] 2. combining the TDS analytical data with the Dietary and Nutritional Survey of British Adults (DNSBA).[6] Using the first method, the mean UK dietary intake of PCDD/Fs fell from 240 pg/TEQ/person/day in 1982 to 69 pg/TEQ/person/day in 1992. With the second method the mean UK dietary intake of PCDD/Fs fell from 250 pg/TEQ/person/day in 1982 to 88 pg/TEQ/person/day in 1992. MAFF suggest that the dietary intakes of dioxins by adult average and high level UK consumers have fallen substantially, and are now below the Tolerable Daily Intake (TDI) of 10 pg/kg body weight per day.

However, there are uncertainties associated with the data used by MAFF. The food groups and consumption/intake surveys used to arrive at the reported downward trend were not the same. For example levels of PCDD/Fs in fruit, vegetables and nuts were not analysed, and fat consumption was markedly lower in the later 1992 TDS.

A remodelling study of MAFF's analytical dioxin intake using the background foods (TDS) data, was undertaken to estimate exposure levels and cumulative intakes in the UK population including those maximally dietarily exposed, - infants and children. Comparisons are made with the adult data from other European countries. This study was conducted in a way not reported to date.

2. Methods/Experimental Design

Methodology

The UK dietary intake/consumption surveys available were examined with regard to reliability and validity of 1). Population sampling; and 2). Dietary intake data.

On the basis of these criteria, with consideration of possible sources of error, the dietary intakes of PCDD/Fs were recalculated with the base data, and expressed in toxic equivalent concentrations (TEQ), using I-TEFs, as used by the Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT).

Significant inadequacies were noted, of which the following are a selected example.

1. Population sampling methods in the 1982 and 1992 NFS's and DNSBA involved multi-stage random probability design sampling of the electoral register subject to alterations of constituency and boundaries, so the TDS 1982 and 1992 assessments have different sampling bases.[2]
Boundary changes not only affect the representativeness of the sample, but alter the variance and shape of the intake distribution, both dietarily[7] and by regional contamination as with Bolsover, Derbyshire.[8]

2. Dietary intake data is not the same as consumption data. In the UK the annual NFS is the major long term food based economic consumption survey of national UK dietary patterns. As it does not record dietary intake, it was not used in this study for intake, although it is part of the TDS from which the background food group samples were analysed. The TDS samples consist of retail food products prepared as for consumption then combined in amounts reflecting their relative importance in the average UK diet as extrapolated from the National Food Survey data supplemented by import and trade statistics. It is based on background foods and does not include vegetables or fruit. The methodology for the TDS is described elsewhere.[1]

Data on individual food intake habits were obtained from :

i) *The Dietary and Nutritional Survey of British Adults 1990* (DNSBA).[6] Over 2000 people, completed a full 7 day weighed dietary record (7DWI) for all food and drink consumed within and outside the home. The subjects ranged from 16 to 64 years of age. A comprehensive range of information was collected, including percentage total fat, dietary supplements and general dietary habits. This survey is considered to be the only nationally representative dietary survey of adults in Britain with a high degree of accuracy due to the use of a more precise food intake methodology [9,10]

ii) *National Diet and Nutrition Survey: children aged 1 and a half to 4 and a half years, Volume 1 Report of the diet and nutrition survey 1995* (NDNS).[11] From a sample of 2101 children, a short 4 day weighed dietary intake was obtained for 1675 children, recording intake both in and outside the home. As with the DNSBA, this survey is considered to be nationally representative ,- of private households.

iii) *Food and Nutrient Intakes of British Infants aged 6-12 months 1992* (FNIBI).[12] The subjects, 488 infants (with a high parental response rate of 81%) were taken from a 'nationally representative' commercial baby panel of over 1800 mothers who kept a 7 day quantitative food record of all food consumed by their infants. Information on sampling procedure to attain national representation is not given.

All the surveys acknowledge under-representation of one parent families and ethnic minorities.
No data of similar quality was available for children aged 5 to 16 years. A joint Department of Health and MAFF survey on Diet and Nutrition in young people aged 4-18 years has just started, but findings will not be available until 1999.

Remodelled data

The TEQ values given in the congener specific analysis of PCDD/F's in samples of fatty foods from the UK total diet study collected in 1992 were recombined with dietary fat intake data according to the informant's sex and age, (income level and region parameters are not reported here) in Microsoft Excel 3.0 spreadsheet format to give sum TEQ dioxin consumption data for adults, children aged 1 1/2 to 4 1/2 years, and infants. See Tables 1, 2, 3.

Statistical analysis

The intake data from the DNSBA were subjected to statistical evaluation, to determine the mean intake and the cumulative intake distribution within the UK population, in relation to age, sex, and bodyweight. Figure 3.

3. Results and Discussion

1) Surveys - criticism and possible sources of error

The following lists examples of sources of error which may significantly underestimate PCDD/F dietary intakes for large sub-groups of the population, and should be accounted for.

i) The recording period of 7 and 4 days was not long enough to record reliable fat intakes, 9 days are required to yield representative data, so there may be substantial under recording of food intake.[6,11,12,13]

ii) Milk and milk products are a major source of the energy foods under-reported. The reduced proportion of milk product intake incorporated into the TDS analysis is lower than expected, and markedly lower than the Dutch value. Figure 2 compares UK/Dutch PCDD/F levels in food commodity groups.[14]

iii) Bread and Cereal products are the only food group that does not follow the indicated general reduction of TEQ levels in all food groups in the MAFF summary[1]. The concentration of PCDD/Fs in cereals is an estimate, not a measured value. However this does not apply to the cereal products category which contains 'hidden' fats. Cereal products are a significant dietary intake route.

iv) Under-representation was observed for younger adults, and low income groups.

v) Fish oils formed 19 % of the supplements taken (DNSBA), particularly among women. Frequency of intake was almost daily. A Dutch dietary intake analysis[14] found that fish oil was responsible for most of the dietary dioxin and PCB contribution in the mixture of vegetable and animal fats and oils (as added by the food industry). Theelen (pers comm) concludes that fish oil use in the food industry is the factor with the greatest uncertainty in the exposure of the general population to PCDD/Fs, as they are also consumed indirectly, through use in the baking and processed food industry, and in fish, meat and egg consumption via animal and fish feed and fish oil rich feed.[15] A recent Spanish analysis of dietary supplement fish oils reported that the daily intake of PCDD/Fs and co planar PCBs via fish oil capsules would be 4.84 pg TEQ/day for an individual weighing 60 kg.[16]

vi) Government Health Policy on the reduction in dietary fat intake may affect consumption patterns. Trends in the lowering of saturated fats have been observed in the DNSBA and the NFS/TDS but a very low level of compliance with Government (COMA) recommendations has been observed.[13] Recent health recommendations specify increasing the -n3 and n-6 fatty acid intake in the form of fish. Fish may be significant dietary sources of dioxins - particularly those farmed on fish oil rich feed.[15]

2) Recalculated results

See Tables 1,2,3. The concentration of dioxin and related compounds for the food categories on a total product basis was calculated by multiplying the percentage total fat in the food item/ category, as recorded in the three dietary survey's, with the concentration TEQ per g fat in the sample from the category representing the food item/ group.

Levels of PCDDs/Fs in fruits and vegetables, vegetable oils and nuts were not included in the 1992 TDS samples, and therefore not in this study. However TEQ values ranging from 0.4 to 0.7 pg/g have been noted in other studies,[17, 18] and have been modelled.[3]

Calculation of individual human daily intake.

For each individual in the dietary intake surveys, the daily intake was calculated by multiplying the amount of the consumed product (gram) with the concentration TEQ (per gram) on a product basis, and summing the TEQs of the different food items consumed. The daily intake per kg body weight for each individual was obtained using the body weight of the individual, as given in the surveys. The average body weight value used was 60 kg.

Distribution of intake

With the statistical model the mean daily dietary intake and the distribution of the TEQ PCDD/F values in relation to age and sex was calculated.

Using the results from the statistical model with the aforementioned parameters, the average cumulative lifetime intakes of dioxin and related compounds was modelled . The results are shown in Figure 3.

No dietary intake data was available for children aged 4 and a half to 16 years (school age), for the same period, therefore the cumulative lifetime intake for this period is modelled on the pre-school and adult dietary intakes.

The cumulative intake model shows a body burden at birth which peaks sharply up to 1 year to almost 80.00 TEQ pg/kg body weight. This falls until 10 years, then steadily rises to 15 TEQ pg/kg body weight at 22 years of age.

Using Dutch food consumption data, R Theelen (pers comm) in his kinetic model for 2,3,7,8 TCDD and related compounds in humans shows a second maximum for young children at five to ten years of age for the total TEQ. If available, the UK dietary intake data for this age group this model for UK intakes would probably indicate that exposure from food is becoming substantially relevant for these children, as well as for young adults.

There is a clear but small difference of dioxin intakes between the sexes. Men have a marginally higher intake due to a greater fat intake. They take a third of their weekly dietary intake outside the home.[6]

3) Implications

An expert group convened by WHO/EURO (1991) has recommended a Tolerable Daily Intake (TDI) of 10 pg/body weight/day for 2,3,7,8-TCDD which is equivalent to an intake of 600 pg TEQ/day for a 60 kg person. This is to be reviewed later in the year. COT recently re-endorsed this TDI, but acknowledged that the daily intake of dioxins could be close to or even above the guideline value.

With this study the estimated average dietary intake of dioxins and furans in the UK is below the TDI recommended by the WHO/EURO expert group (1991) for adults at 2.93 pg TEQ/kg. body weight/day, or 175.5 pg/TEQ/person/day, in contrast to MAFF estimates of 69 and 88 pg/TEQ/person/day,[2] and a recently modelled value of 61 pg/TEQ/person/day, based on the US EPA food chain exposure model and using NFS and TDS data.[3] The lower figures do not include food eaten outside the home.

The levels for young children exceed the TDI, ranging from 54.19 TEQ/kg b.w./day at 6 months of age down to .0.25 pg TEQ/kg b.w./day at 4 1/2 years. See Table. 3

This compares favourably with a small German PCDD/F balance study, where the measured levels were 38.3 and 49.0 pg/kg b.w/day at 5 months for 2 breast fed infants, dropping to 3.6 and 1.3 pg/kg b.w/day at 10+ months, and 2 months after weaning. Levels after weaning for 2 breastfed and 2 formula fed infants aged 10 to 13 months were 1.3-3.6 pg kg b.w/day.[19]

4. Conclusion

Including breast milk intake, young children in the UK are well above the TDI for PCDDs/Fs, and adult intake is at least twice that estimated by MAFF.

Comparison with recent European studies based on consumption /intake data from variable sources (Figure 1) indicates that this study gives the highest representative estimate of average mean PCDD/F dietary intakes. The relative contribution of milk fat to the total diet is unusually low compared with the Dutch levels, (see Figure 2), but may be explained by under-reporting.

Sections of the population with a higher dietary exposure include those (e.g. children) with an above average intake of milk products, offal's, oils ands fats, fish and cereal products .

Processed foods are a significant route for human dioxin exposure because animal and fish oils are used extensively in the European food industry. Fish oils have been noted to determine approximately 90% of the quantity of dioxins, furans and PCBs in Dutch industrial fats.[14]

Contrary to other estimates[2,3] there does not appear to be a clear downward trend in dietary intake of PCDD/Fs in the UK, and it cannot be concluded that current emission controls on the combustion processes which cause PCDD/Fs to form, are sufficient.

Infant and child cumulative intake may even out over time (see Figure 3), but such raised levels of intake at crucial times of development may represent a missed opportunity with a lasting effect as seen with later developmental problems or the development of childhood cancer.[20]

5. Acknowledgements

We would like to thank FoE UK and BERG for help with funding.

6. Main literature cited

1. Wright, C., & Startin, J.R. *Ministry of Agriculture, Fisheries and Food Report No. FD94/200.* July **1995,** MAFF.
2. Wearne, S.J., Harrison, N., de Gem, M., Startin, J. R., Wright, C., Kelly, M., Robinson, C. White, S., Hardy. D., Edinburgh. V. *Organohalogen Compounds,* 1996, 30, 1-5.
3. Eduljee, G.H.., Gair, A.J. *The Science of the Total Environment* .**1996** 211-229
4. Ministry of Agriculture Fisheries and Food *National Food Survey* HMSO , London **1982**
5. Ministry of Agriculture Fisheries and Food *National Food Survey* HMSO. London **1992**
6. Gregory. J., Foster, K., Tyler, H., and Wiseman, M. *Dietary and Nutritional Survey of British Adults.* HMSO, London **1990**.
7. Pryer, J., Brunner. E. and Marmot, M. *J. Epidemiol. Community Health.* **1994** 48, 543-8.
8. Ministry of Agriculture Fisheries and Food *Food Surveillance Information Sheet* 100 HMSO. London **1997**
9. Brunner, E.J., Sharma, A., Pryer J.A.. Elliot. P.. and Marmot. M.G. *WHO European 3rd Nutritional Epidemiology Conference, Berlin, 224.* **1992**
10. Bingham. S.A. (1991) *Ann. Nutr. Metab.* **1991,** 35, 117-127.
11. Gregory, J. R.. Collins, D. L., Davies, P.S.W., Hughes, J.M., Clarke, P.C. *National diet and nutrition survey: children aged 1 1/2 to 4 1/2 years Volume 1: Report of the diet and nutrition survey.* London HMSO. **1995.**
12. Mills, A., and Tyler, H. *Food and Nutrient Intakes of British Infants aged 6-12 months.* London HMSO. **1995.**
13. Pryer. J., Brunner, E., Elliot, P., Nichols, R., Dimond, H.. and Marmot, M. *Eur J of Clin Nutr.* **1995.** 49. 718-28.
14. Theelen, R.M.C., Liem, A.K.D., Slob, W., and Van Vijnen. J.H. *Chemosphere.* **1993,** 27, 9, 1625-35.
15. Jacobs, M.N.. and Johnston, P.A. *International Journal of Environment and Pollution.* **1997**. in press.
16. Jimènez, B., Wright, C., Kelly, M., and Startin, J.R. *Chemosphere.* **1996,** 32. 451-7.
17. Fürst. P., Fürst, C., Groebel, W. *Chemosphere.* **1990,** 20, 787-792
18. Birmingham, B., Thorpe, B., Frank, R., Clement, R., Tosine, H., Fleming, G., Ashman, J., Wheeler, J., Ripley. B.D., Ryan, J.J. *Chemosphere.* **1989,** 19, 504-512
19. Abraham, K., Knoll. A., Ende, M., Päpke, O., Helge, H. *Pediatr Res.* **1996,** 40, 5, 671-679 .
20. Knox, E.G., & Gilman, E.A. *J. Epidemiol. Community Health.* **1997**, 51:151-159
21. Jimènez, B., Hernàndez, L.M., Gonzàlez, M.J., Elijarrat, E., Rivera, J. *Organohalogen Compounds.* **1996,** 30. 131-136

Table 1. Male/Female dioxin intake (based on fat intake)

Commodity	Dioxin* ng/kg in..	Fat Intake**, g/d Male	Fat Intake**, g/d Female	Dioxin intake Male pg/d	Dioxin intake Female pg/d
Bread	0.03				
Cereal prod.	0.17	18.9	14.9	3.21	2.53
Carcass meat	0.13				
Offal's	0.59				
Meat prod.	0.08	26.9	16.2	2.15	1.30
Poultry	0.13				
Fish	0.21	2.9	2.0	0.61	0.42
Oils and fats	0.20	16.6	11.8	3.32	2.36
Eggs	0.17	4.2	3.0	0.71	0.51
Milk	0.06				
Milk prod.	0.16	15.0	11.8	2.40	1.89
Vegetables	0.00	11.4	8.0		
TOTAL:		**95.9**	**67.7**	**12.41**	**9.01**
Intake, pg/kg bodywt/day				**0.21**	**0.15**
Cumulative intake, pg/kg body wt/day				**1.96**	**1.43**

* derived from data supplied by MAFF using 1992 TDS analytical values

** derived from data supplied by the Department of Health/MAFF

Fat intake from outside home...	Male	Female
Intake of fat, g/day	102.30	73.50
% of non-domestic intake	31.8%	24.7%
Intake outside home, g/day	32.82	25.44
Dioxin in oils & fats, ng/kg	0.20	0.20
Dioxin intake, pg/day	6.56	5.09
Intake, pg/kg bodywt./day	0.11	0.08
Cumulative intake, pg/kg	1.04	0.81

Total domestic/non-domestic intake from fat pg/kg bodywt./day	**0.32**	**0.23**
Total cumulative intake from fat, pg/kg body weight/day	**3.00**	**2.23**

In some instances the food commodities have been grouped together to accommodate the survey data with the analytical data.

Table 2. Dioxin intake by age

Commodity	Dioxin* ng/kg, in..	Food intake**, g/day, by age... 16-24	25-34	35-49	50-64	Dioxin intake, g/day, by age... 16-24	25-34	35-49	50-64
Bread	0.03	165.9	157.0	160.6	168.3	4.98	4.71	4.82	5.05
Cereal prod.	0.17	265.1	219.7	263.7	234.9	45.07	37.35	44.83	39.93
Carcass meat	0.13	115.4	130.6	116.6	117.7	15.01	16.97	15.15	15.30
Offal's	0.59	16.7	17.6	17.1	17.0	9.86	10.37	10.11	10.03
Meat prod.	0.08	135.7	109.4	100.9	94.0	10.86	8.75	8.07	7.52
Poultry	0.13	61.0	58.6	52.4	52.7	7.93	7.61	6.82	6.85
Fish	0.21	82.4	78.1	86.6	85.9	17.31	16.41	18.18	18.03
Oils and fats	0.20	60.4	60.4	65.4	75.7	12.09	12.09	13.09	15.14
Eggs	0.17	28.7	29.6	28.7	27.3	4.88	5.03	4.88	4.64
Milk	0.06	465.9	475.9	525.6	526.1	27.95	28.55	31.53	31.57
Milk prod.	0.16	142.6	124.0	140.1	135.0	22.81	19.84	22.42	21.60
Vegetables	0.00								
TOTAL:		**1539.9**	**1460.9**	**1557.7**	**1534.6**	**178.74**	**167.69**	**179.91**	**175.66**
		Intake, pg/kg body weight /day				**2.98**	**2.79**	**3.00**	**2.93**
		Cumulative intake pg/kg bodywt/day				**28.30**	**26.55**	**28.49**	**27.81**

* derived from data supplied by MAFF

** derived from data supplied by the Department of Health/MAFF

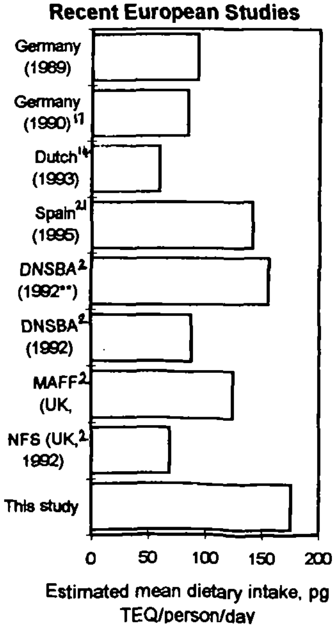
Table 3. Children's dioxin intake (based on fat intake)

Commodity	Dioxin* ng/kg, in..	Fat intake**, g/day, by age... 6 to 9 mths	9 to 12 mts	1.5 to 2.5 yrs	2.5 to 3.5 yrs	3.5 to 4.5 yrs	Dioxin intake, g/day, by age... 6 to 9 mts	9 to 12 mts	1.5 to 2.5 yrs	2.5 to 3.5 yrs	3.5 to 4.5 yrs
Bread	0.03	0.9	3.0	3.6	4.2	4.7	0.03	0.09	0.11	0.13	0.14
Cereal prod.	0.17	1.4	9.0	4.0	5.3	5.6	0.24	1.53	0.68	0.90	0.95
Carcass meat	0.13										
Offal's	0.59										
Meat prod.	0.08	1.1	6.0	5.8	6.6	7.5	0.09	0.48	0.46	0.53	0.60
Poultry	0.13	0.2	1.0	0.6	0.8	0.9	0.03	0.13	0.08	0.10	0.12
Fish	0.21	0.2	1.0	0.9	0.9	1.0	0.04	0.21	0.19	0.19	0.21
Oils and fats	0.20	1.2	2.9	3.5	4.5	5.0	0.24	0.58	0.70	0.90	1.00
Eggs	0.17	1.2	2.2	1.1	1.3	1.4	0.20	0.37	0.19	0.22	0.23
Milk	0.06	8.3	14.7	11.4	9.1	7.9	0.50	0.88	0.68	0.55	0.47
Milk prod.	0.16	0.7	1.5	3.4	2.8	3.0	0.11	0.24	0.54	0.45	0.47
Vegetables	0.00	0.2	3.0	5.1	6.4	7.1	0.00	0.00	0.00	0.00	0.00
Formula milk	2.10***	3.0	0.8				6.30	1.68	0.00	0.00	0.00
Breast milk	22.00	9.5	3.2				209.00	70.40	0.00	0.00	0.00
TOTAL:		**27.9**	**48.3**	**39.4**	**41.9**	**43.9**	**216.78**	**76.60**	**3.63**	**3.96**	**4.19**
			Intake, pg/kg body weight/day				54.19	12.77	0.29	0.28	0.25
			Cumulative intake pg/kg body wt.				514.84	121.28	2.75	2.63	2.41

* derived from data supplied by MAFF

** derived from data supplied by the Department of Health/MAFF *** Abraham K. et al. 1996

Fig. 1. Comparison of Recent European Studies



Year denotes year of sample.

** maximum exposed groups

Fig. 2. Comparison of UK/Dutch PCDD/F Levels in Foodstuffs (in fat)

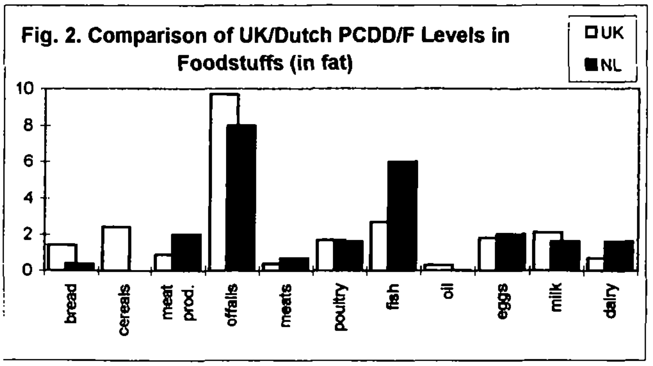
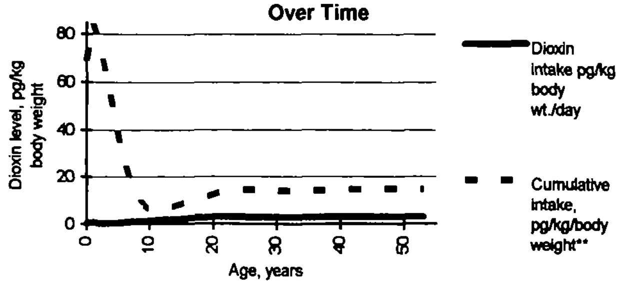


Fig. 3. UK Dioxin Intake and Cumulative Intake Over Time



**assuming theoretical 18 year life for sum TEQ PCDD/Fs.
Half life levels for individual congeners have been reported to range from 7.2 to 19.6 yr (Flesch-Janys et al 1996)