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Report of experts participating in Task 3.2.12

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COLLECTION OF OCCURRENCE DATA ON POLYCYCLIC AROMATIC HYDROCARBONS IN FOOD

Directorate-General Health and Consumer Protection

TASK 3.2.12 "COLLECTION OF OCCURRENCE DATA ON POLYCYCLIC AROMATIC HYDROCARBONS IN FOOD"

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FOREWORD

Council Directive 93/5/EEC "on the assistance to the Commission and co-operation by the Member States in the scientific examination of questions relating to food" was updated on 25 February 1993. It lays down a procedure whereby Member States of the European Union can focus their scientific resources in a co-ordinated manner on problems facing the European Commission in the area of food. The individual tasks to be undertaken are agreed in consultation with the Member States who also determine in which tasks they wish to participate and the extent of their participation. Directive 93/5/EEC requires that an inventory of tasks be published at least every six months. This publication, which takes the form of a Commission Decision, specifies the participating Member States that provides co-ordination and the time limit for the completion of the task.

In general terms, tasks undertaken under scientific co-operation are designed to provide a factual basis to support a Commission action in the area of food. Such support may involve the provision of information as may be required, for example, by the Scientific Committee for Food (SCF) for its evaluation and advisory work or by the Commission's own services for the development of Community action.

The tasks themselves are carried out by a group of experts nominated by the National Authorities responsible for Scientific Co-operation in the Member States (the National Designated Authorities).

Although the scope of reports generated under the scientific co-operation procedure is restricted to essentially factual matters, presentation of inherently complex information without some reasoned interpretation and summary by specialists would be of limited value and even open to misleading conclusions. Such interpretation necessarily involves a degree of expert judgement.

It is therefore stressed that the interpretation and views expressed in this report are not necessarily those of the participating Member States or those of the European Commission.

Council regulation EEC 315/93 of 8 February 1993 provides the legal framework for establishing maximum levels for food contaminants at Community level,
Council Directive 93/5 EEC provides the legal framework on assistance and cooperation by the Member states in the scientific examination of question relating to food,
Commission decision 94/548 laid down rules on the administrative management of cooperation in the scientific examination of questions relating to food.

Following the advice of the EU Scientific Committee on Food of December 2002, the Commission is considering proposing the establishment of maximum levels for Polycyclic Aromatic Hydrocarbons (PAH) in food at Community level, based on the legal framework of Council Regulation EEC 315/93 of 8 February 1993 (EEC, 1993). It therefore seeks data for these considerations.

The Commission decision of 26 October 2001 appointed the AFSSA as coordinating institute for the SCOOP Task 3.2.12. to collate data on PAH in food.

INTRODUCTION

The objectives of the SCOOP Task were to collect and collate occurrence data on PAH in a range of foodstuffs These particular requirements arose because several PAH have been recognized as probable human carcinogen (SCF, 2002).

Initially, the purpose was to gather the available occurrence data on PAH in food. Due to issues arising during the last past years about high level of contamination in specific foodstuffs (e.g. olive pomace oil), the possibility also to include intake levels of PAH was considered in order to approximate exposure levels to PAH from food consumption. The coordinator and the participants of the task agreed that it would be possible, in some cases, to provide data on dietary intake and exposure to these compounds in individual Member State.

Thirteen countries have participated and provided information : Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland (Republic of), Italy, The Netherlands, Norway, Portugal, Spain and the U.K. Details of participants are provided in Annex 5.

Basically, participants were asked to provide information on the exposure of the population to PAH in their country through the elaboration of the following categories of data:

- ✓ Occurrence data in food and beverages, including those from different methodological approaches (i.e. total diet, duplicate test portion),
- ✓ Consumption data,
- ✓ Best estimate of dietary intake.

In addition, the following technical information were deemed relevant:

- ✓ Sampling procedures,
- ✓ Quality assurance of analytical data.

Before beginning the collection of data, the SCOOP Task participants have developed and agreed upon the best format for this purpose.

PAH originate from different sources; they constitute a family of compounds with diverse chemical and biological properties (Annex 1). After discussion with the SCF WG and the European Commission services, a number of considerations were gathered for providing the most convenient and useful information as regard both handling, reliability and relevance of the expected considerable amount of data available.

The following requirements were identified :

- ✓ to identify which foods (either raw or processed) are the main sources of PAH in the "regular" diet.
- ✓ to identify which foods may be the most likely target of high contamination. This can be during specific episodes of contamination, either limited such as consecutive to an accident, or long lasting in the case of inadequate processing or of polluting emissions from an industrial plant. It can also be the consequence of a regular preparation such as cooking, frying and smoking.
- ✓ as far as possible to allow calculation of the background exposure to PAH, (thus excluding accidental exposure to abnormally high levels of PAH).

List of relevant PAH.

The issues of "the setting criteria" and of "the nature of" relevance have been discussed. Several criteria can be used :

- ✓ Concentrations in foods: e.g. PAH found in large amount in foods.
- ✓ Frequency of occurrence (detection) in foods.
- ✓ Origin : natural or anthropogenic, taking into account that the presence of "man made" PAH in food can be derived (can the "presence" be "derived" …from several types of processes. PAH are formed during regular cooking or smoking, or not intentionally such as those PAH produced in overcooked fat meat and fishes or, contaminating the environment and consequently, the foods.
- ✓ Hazard : e.g. PAH which represent the highest toxicological concern. Preferentially, the type of effect and the effective dose should be known. Therefore, all PAH that have been allocated carcinogenic TEF should be included (if present in the diet).

It was decided to concentrate on the 16 PAH that are in the so-called EPA-list, then possibly extend to the 33 PAH that have been studied in the 1998 IPCS report. (The SCOOP data was mostly gathered before the opinion of the SCF was finalised in December 2002, hence the focus of SCOOP was not on the 15 PAH considered to be of most concern by SCF.) Also, it was decided not to limit to the PAHs which can be, to different extents, of cancer risk, but to collect all available data, even including as a matter of caution, alkylated PAH for which the toxicological knowledge is very poor. It was decided to register any available data that were considered as sufficiently robust. Knowledge of the reason why these PAH have been measured was taken into account where data could otherwise give rise to irrelevant values leading to wrong calculation of mean intake.

List of relevant foods

On the basis of the available literature, it was deemed necessary to identify and select the foods which are the most potent sources of PAH in the diet.
Due to their conditions of formation in foods which are almost unique to PAH, the following issues were discussed:

- ✓ - Are there data available in foods under various stages of preparation: as sold (raw), as eaten (after cooking), unprocessed, frozen, "ready to eat" foods (microwavable).
- ✓ - What is the usefulness of indications on the regional origin of foods : not imported (domestic, in house), or imported (country of origin).

Another set of criteria have been deemed worth discussing, on the background that there are known confounding factors that do notably influence the reliability of data.

Methodological matters.
Sampling

- ✓ Origin, handling, storage conditions, processing (extraction procedure, separation, clean up).

Analytical procedures for identification and quantification

- ✓ Type of analytical methodology used: gas- (GC), high performance liquid-(HPLC), or thin layer-(TLC) chromatography together with the type of detection: flame ionisation (FID), mass spectrometry (MS), ultra violet (UV) or fluorescence (FLD).

- ✓ Limits of detection (LOD) and of quantification (LOQ) of the analytical method used,
- ✓ Uncertainty (%).

Quality Assurance information:

- ✓ Use of a standard: internal, external (participation in proficiency test schemes),
- ✓ Accreditation of the laboratory if relevant.

Given the complexity of the data requirements, a dedicated framework (template) was designed specifically for this SCOOP Task. It was presented, discussed, modified and made available after final agreement, to all participating members.

Consequently, PAH occurrence data have been collated into an electronic database (Excel spreadsheet), which may be used as a source of various information (Annexes 2 and 3).

Given the wide variations among member states in:

- ✓ The pattern of monitored compounds,
- ✓ The origin: regular survey, target/random monitoring, total diet study (examples of differences are shown in next section: monitoring procedures), and expression of data: use of Toxicological Equivalent Factors (TEF), wet or dry weigh basis.
- ✓ The types of sampling and of analytical methodology and procedures,
- ✓ The families (number and composition) of foods that were considered for determinations,

it was decided, after discussion with the Commission services, that the report will be made of an executive summary giving the main observations, and drawing the main conclusions and of annexes in which the database will be presented, as usual, country by country (annexes 6, 7 and 8).

Efforts have been made to present data in certain areas with a higher degree of detail where results have been produced with a level of confidence high enough to allow comparison. This is particularly the case of benzo(*a*)pyrene : BaP, which is the member of the PAH family the most frequently assayed (99%).

Mean values for BaP have been calculated for several food groups (tables 4-17 pp 14-20). Given Germany is the only MS to have provided data for waters, these should be considered as non representative of exposure of european citizens. However, it seemed relevant to include the data in the report due to the significant amount of knowledge they represent.

EXAMPLES OF MONITORING PROCEDURES.

Germany.

The German food control is not a centralised system.

Official food control or random monitoring.

The German food control is a system independently managed by every German Länders. The number of food samples per inhabitants is fixed in every Länders. The type of sampling is random. The authorities or laboratories of the Länders regulate which parameter or substance were investigated in each food sample.

Target monitoring in contaminated areas / of suspected contaminated foodstuffs.

Also a monitoring plan for Germany exists. Every year the authorities of the food control lay down which substance had to be examined in a fix number of food samples in regulate food groups. The target monitoring contains this monitoring plan and also samples of research projects of the accredited national laboratories of food control or federal laboratories of research.

Suspicion control.

The authorities have to take samples if somebody makes a complaint by the authorities of the Länders.

United Kingdom.

Total Diet Study (TDS) samples consist of retail food product prepared as for consumption and then combined into composite samples in amounts reflecting their relative importance in the typical UK diet. Each composite sample represents a defined food group. A total of 2000 retail samples are collected each year at fortnightly intervals from 24 locations in the UK and pooled into 20 single samples (one for each food group) for analysis. All TDS samples are stored under stable conditions.

Norway.

General comments on selection of data/ Description of the Norwegian sampling procedure

Samples were taken in the year of 2002. Most of the samples were of processed food, but a few consisted of raw commodities, such as pork chops and chicken. For most meat and fish products three individual samples were taken from three of the major food chains. Samples were also taken of different kinds of bread (wheat- and whole wheat), biscuits and breakfast cereals. Ten individual samples of olive oil and 6 samples of sunflower-, corn- and rape oil were taken. The sampling did not cover all foods that are considered to be a part of a Norwegian diet. For instance, no sampling of fruits and vegetables were made.

France.

978 contamination data were collected by DGAL[1] and the DGCCRF[2] as part of targeted or random inspection plans conducted in 1999 and 2001 and surveillance plans conducted between 2000 and 2002 following the wreck of the oil tanker Erika. The samples concern foods as consumed, with no assumptions as to preparation prior to consumption.

[1] DGAL: Direction générale de l'alimentation

[2] DGCCRF: Direction générale de la concurrence, de la consommation et de la répression des fraudes.

RESULTS.

1 PAH OCCURRENCE IN FOOD.

Concerning the occurrence data on PAH in foodstuffs a total of 8861 samples have been collected from 14 countries (13 Member States and Norway).

The assays were performed during a period from 1992 until 2003.

A list of 44 food groups was initially proposed in the questionnaire but after collation of the occurrence data, it appeared that an additional group named "Additives" mostly composed of liquid smoke flavouring samples was needed. Thus 45 food groups were considered (table 1).

Concerning the food group "Waters", it was decided in agreement with the Commission services, to not include the data on tap water in the intake calculation. However, as stated above, because they represent a significant input as regards knowledge, it was decided to present the occurrence data.

As a first outcome, the distribution is unbalanced given that 5 food groups gathered more than 80% of the samples:

- Sausages & ham (2358 samples, 27%)
- Vegetal oils (2110 samples, 24%)
- Fish / fish products (1173 samples, 13%)
- Waters (excluding tap water) (1004 samples, 11%)
- Meat (519 samples, 6%)

Table 1: Samples distribution within food groups.

Food groups	Number of samples
Additives	83
alcoholic beverages excluding beer & wine	1
beer & wine	22
Biscuits	12
bread / rolls	**103**
breakfast cereals	12
butter & animal fats	6
cereals (others)	63
Cheese	41
chocolate (chocolate & cocoa products)	16
coffee (green bean, filter, instant)	32
crisps & ready-to-eat savouries (salted snacks & seeds)	11
Crustaceans	24
dairy products	1
dried fruits	**158**
Eggs	4
fish / fish products	**1179**
fries / chips	2
fruit juices	2
Fruits	95
hot drinks (tea, infusion excluding cocoa powder)	57
Margarine	34
meat	**519**
Milk	1
mixed dishes	20
Molluscs	**300**
nuts & oleaginous grains	30
Offals	2
Others	12
pastas / rice	16
pastry goods	8
pizzas & quiches	20
Potatoes	17
poultry & game	28
products for special nutritional use (infant & dietetic formulae)	22
sausages & ham	**2358**
soft drinks excluding fruit juices	2
spices/sauces/condiments	51
sugar & by-products including honey	16
vegetables (canned)	1
vegetables (leaf)	64
vegetables (others)	87
vegetables (root excluding potatoes)	24
vegetal oils	**2110**
waters (mineral, source, tap)	**1195**
Total	**8861**

Regarding PAH determined, data for 31 different compounds (list below) were collected with a highly variable pattern of determination among countries (table 2) :

Parent PAH :

- Acenaphthene
- Acenaphthylene
- Anthanthrene
- Anthracene
- Benz[*a*]anthracene
- Benzo[*b*]fluoranthene
- Benzo[*j*]fluoranthene
- Benzo[*k*]fluoranthene
- Benzo[*ghi*]perylene
- Benzo[*a*]pyrene
- Benzo[*e*]pyrene
- Chrysene
- Cyclopenta[*cd*]pyrene
- Dibenz[*a,h*]anthracene
- Fluoranthene
- Fluorene
- Indeno[1,2,3-*cd*]pyrene
- Naphthalene
- Perylene
- Phenanthrene
- Pyrene
- Triphenylene

Alkyl derivatives:

- 1-Methylnaphthalene
- 2-Methylnaphthalene
- 2,6-Dimethylnaphthalene
- 2,3,5-Trimethylnaphthalene
- 1-Methylphenanthrene
- C2- Phenanthrenes

Heterocyclic aromatic hydrocarbons:

- Benzo[*b*]naphto[2,1-*d*]thiophene
- Dibenzothiophene
- C1-dibenzothiophenes

Table 2 : PAH distribution by country (number of samples analysed for each individual PAH).

PAH analysed	Austria	Belgium	Denmark	Finland	France	Germany	Greece	Ireland	Italy	Netherlands	Norway	Portugal	U.K.	Spain	Total
Number of samples	1137	60	98	341	1177	4917	198	156	307	38	138	92	53	149	8861
Acenaphtene		60	34	341		701			116	38	138	92	53	4	1577
Acenaphthylene		60	98	341		592			116		138	92	53		1490
Anthracene		60	34	341	395	793	146		203	38	138	92	53	4	2297
benz(*a*)anthracene		60	34	341	1177	979	175		249	38	138	92	53	98	3434
benzo(*b*)fluoranthene					1177	2024	175		222	38		65	53	87	3841
benzo(*k*)fluoranthene		60	64		1068	2024	85		220	38		65	53	87	3764
benzo(*gh*i)perylene		60	98	341	1068	2134	85		256	37	137	59	53	71	4399
benzo(*a*)pyrene	**1137**	**60**	**98**	**341**	**1177**	**4876**	**198**	**156**	**288**	**38**	**138**	**88**	**53**	**149**	**8797**
chrysene		60			690	811	58		201	38		40	53	8	1959
dibenz(*a,h*)anthracene		60	34	341	1177	1101	85		223	37	138	32	53	102	3383
fluoranthene		60	98	341	636	1865	154		207	38	138	40	53	15	3645
fluorene		60	34	341	12	704			176	38	138	40	53		1596
indeno(1,2,3-*cd*)pyrene		60	98	341	945	2103	83		216	24	138	56	53	87	4204
naphtalene		60	34	341	320	690			164	38	138	44		4	1833
phenanthrene		60	34	341	428	810	152		203	38	138	44	53	11	2312
pyrene		60	34	341	316	839	151		206	38	138	44	53	15	2235
benzo(e)pyrene			34	341		109			124		20	31	20	71	750
benzo (*b*)+(*j*) fluoranthene		60	64												124
triphenylene + chrysene			34	341					20		138				533
benzo(*j*)fluoranthene					60										60
benzo (*a*)+(*k*) fluoranthene						107									107
sum (FL,BaP,BbF,BkF,BeP,IP)						31									31
benzo (*b*)+(*k*) fluoranthene						34									34
Benzo (*b*)+(*j*)+(*k*) fluoranthene				341					35		138				514
2-methylnaphthalene			34								20				54
1-methylnaphthalene			34								20				54
2,6-Dimethylnaphthalene			34								20				54
2,3,5-Trimethylnaphthalene			34								20				54
1-Methylphenanthrene			34								20				54
Perylene			34								20				54
Anthanthrene													20		20
benzo(b)naphtho(2,1-d)thiophene													20		20
cyclopenta(cd)pyrene													20		20
Dibenzothiophene			34												34
C1-dibenzothiophenes			34												34
C2-phenanthrenes			34												34

Table 3 : Determination of PAH in samples (from 1 to 19 different PAH determined in separate samples).

PAH analysed	Total	% of samples analysed	Genotoxicity	Carcinogenicity
Number of samples	8861			
Acenaphthene	1577	17.8%	(?)	?
Acenaphthylene	1490	16.8%	(?)	no study
Anthracene	2297	25.9%	-	-
benz(*a*)anthracene	3434	38.8%	+	+
benzo(*b*)fluoranthene	3841	43.3%	+	+
benzo(*k*)fluoranthene	3764	42.5%	+	+
benzo(*ghi*)perylene	4399	49.6%	+	-
benzo(*a*)pyrene	**8797**	**99.3%**	**+**	**+**
chrysene	1959	22.1%	+	+
dibenz(*a,h*)anthracene	3383	38.2%	+	+
fluoranthene	3645	41.1%	+	(+)
fluorene	1596	18.0%	-	-
indeno(1,2,3-*cd*)pyrene	4204	47.4%	+	+
naphtalene	1833	20.7%	-	(?)
phenanthrene	2312	26.1%	(?)	(?)
pyrene	2235	25.2%	(?)	(?)
benzo(*e*)pyrene	750	8.5%	+	?
benzo (*b*)+(*j*) fluoranthene	124	1.4%	mixture	mixture
triphenylene + chrysene	533	6.0%	mixture	mixture
benzo(*j*)fluoranthene	60	0.7%	+	+
benzo (*a*)+(*k*) fluoranthene	107	1.2%	mixture	mixture
sum (FL,BaP,BbFA,BkFA,BeP,IP)	31	0.3%	mixture	mixture
benzo (*b*)+(*k*) fluoranthene	34	0.4%	mixture	mixture
Benzo (*b*)+(*j*)+(*k*) fluoranthene	514	5.8%	mixture	mixture
2-methylnaphthalene	54	0.6%		
1-methylnaphthalene	54	0.6%		
2,6-Dimethylnaphthalene	54	0.6%		
2,3,5-Trimethylnaphthalene	54	0.6%		
1-Methylphenanthrene	54	0.6%		
Perylene	54	0.6%		
Anthanthrene	20	0.2%	(+)	+
benzo(*b*)naphto(2,1-d)thiophene	20	0.2%		
cyclopenta(*cd*)pyrene	20	0.2%	+	+
Dibenzothiophene	34	0.4%		
C1-dibenzothiophenes	34	0.4%		
C2-phenanthrenes	34	0.4%		

+ : positive ? : questionable () results from a limited database

From : IPCS, EHC 2002.

The expression of results was different depending on the laboratory. Some were given as : Not Detected (below the limit of detection : LOD), and some as : Not Quantified (below the limit of quantification : LOQ).
Considering that these limits are highly dependent on the analytical methodology used, these differences in LOD / LOQ together with the large amount of results below LOQ, made the interpretation and use of data difficult.

As a result, because values such as : <LOQ or <LOD, do not necessarily mean that PAH are absent from the sample, it was decided to assume a medium value to individual results in order to calculate best estimate mean values to be used in the intake analyses.

Therefore, the samples below the LOQ or LOD were assigned a value of LOD/2 or LOQ/2.

Mean values for BaP have been calculated for several food groups (tables 4-17).

Table 4: BaP occurrence in the food group "Additives".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50ppb	Nb samples 50-100ppb	Nb samples >100 ppb
Additive: smoke flavouring	30	11	12	17	11	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Additive: smoke flavouring	81	2.60	1.00	4.18	13.20	16.96

Table 5: BaP occurrence in the food group "Bread & rolls".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
Bread & rolls	101	0	1	0	1	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Bread & rolls	103	0.16	0.05	0.55	0.30	1.48

Table 6: BaP occurrence in the food group "Cereals".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples > 100 ppb
Cereals	56	5	2	0	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Cereals	63	0.16	0.05	0.31	0.88	1.39

Table 7: BaP occurrence in the food group "Cheese".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
Cheese	38	2	1	0	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Cheese	41	0.19	0.05	0.29	0.72	1.24

This food group includes 41 samples out of which 22 had been smoked using various procedures (traditional, liquid flavouring).

Table 8: BaP occurrence in the food group "Dried fruits".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
Dried fruits	103	4	5	5	14	5	6	16

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Dried fruits	158	48.10	0.25	5.30	447.20	670.19

Table 9: BaP occurrence in the food group "Fishery products".

Fishery products were split into sub-groups : fishes, molluscs, crustaceans (fresh or processed)

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food subgroups	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
fresh crustaceans	21	0	0	0	1	0	0	0
fresh molluscs	129	61	19	29	18	6	1	1
fresh fishes	429	13	7	2	3	0	0	0
canned fishes in oil	111	19	18	14	9	0	0	0
smoked fishes (method unknown)	97	8	9	7	5	0	1	0
smoked fishes (liquid flavouring)	12	0	0	0	0	0	0	0
Smoked fishes (traditional method)	177	12	1	6	3	5	6	3

unit in µg/kg wet weight

food subgroup	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
fresh crustaceans	22	0.64	0.36	1.42	0.45	5.73
fresh molluscs	264	3.09	0.53	10.90	16.05	34.15
fresh fishes	454	0.21	0.08	0.71	0.53	2.39
canned fishes in oil	171	1.11	0.25	2.46	5.05	13.60
smoked fishes (method unknown)	127	1.43	0.20	7.15	4.10	15.27
smoked fishes (liquid flavouring)	12	0.03	0.03	0.02	0.08	0.08
smoked fishes (traditional method)	213	5.28	0.05	21.70	37.68	116.66

fresh crustaceans:
21 samples were below LOQ, 1 sample was around 8 ppb (living pink shrimp).
fresh molluscs:
264 samples available, 23 samples from monitoring in contaminated area (Baltic Carrier Oil spill). For these 23 samples, the BaP concentration ranged between 2 and 23 ppb.
fresh fishes:
454 samples available;11 samples were from monitoring in contaminated area (Baltic Carrier Oil spill) for these 11 samples BaP concentration ranged between 1 and 9 ppb.
smoked fishes (unknown method):
127 samples available with 1 sample from a targeted monitoring of suspected contaminated foodstuffs (smoked sardine) containing around 80 ppb BaP.
smoked fishes (liquid flavouring):
the 12 samples were below 0.1 ppb.
smoked fishes (traditional method):
from the 213 samples 12 were from monitoring of suspected contaminated foods. BaP ranged 12-140 ppb. 14 samples were > 20 ppb, 9 samples >50 ppb, and 3 >100 ppb.

Table 10: BaP occurrence in the food group "Fruits".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples > 100 ppb
Fruits	92	2	0	1	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Fruits	95	0.07	0.02	0.23	0.15	0.95

Table 11: BaP occurrence in the food group "Margarine".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
Margarine	32	0	2	0	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Margarine	34	0.18	0.10	0.33	0.78	1.47

Table 12: BaP occurrence in the food group "Nuts & oleaginous grains".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-10 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples > 100 ppb
nuts & oleaginous grains	28	1	0	1	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
nuts & oleaginous grains	30	0.27	0.10	0.89	0.52	3.75

Table 13: BaP occurrence in the food group “Smoked products”.

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples > 100 ppb
smoked products (method unknown)	123	2	8	2	6	3	1	0
smoked products (liquid flavouring)	198	0	0	0	0	0	0	0
smoked products (traditional method)	670	63	63	97	88	29	9	4

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
smoked products (method unknown)	145	1.74	0.06	6.70	9.70	35.60
smoked products (liquid flavouring)	198	0.023	0.015	0.027	0.06	0.11
smoked products (traditional method)	1023	3.27	0.13	11.70	15.63	54.86

smoked products (method unknown):
145 samples available (23 meat samples, 6 poultry & game, 116 sausages & ham), 4 samples were > 20 ppb.

smoked products (liquid flavouring):
198 samples available (13 meat samples, 6 poultry & game, 179 sausages & ham). The mean BaP contamination was 0.02 ppb.

smoked products (traditional method):
1023 samples available (12 meat samples, 7 poultry & game, 1004 sausages & ham), 42 samples were > 20 ppb, 13 samples > 50 ppb, 4 samples > 100 ppb. An explanation for the high levels was not achievable due to tracking difficulties of the samples.

Table 14: BaP occurrence in the food group "Spices/sauces/condiments".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb Samples 50-100ppb	Nb samples > 100 ppb
spices/sauces/condiments	29	3	11	4	3	1	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
spices/sauces/condiments	51	2.16	0.39	6.93	6.75	32.65

Table 15: BaP occurrence in the food group "Vegetables".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
Vegetables (leaf)	62	0	0	2	0	0	0	0
Vegetables (others)	71	5	3	5	3	0	0	0
vegetables (others) raw	45	0	0	3	2	0	0	0
vegetables (others) processed	26	5	3	2	1	0	0	0
Vegetables (root)	22	0	2	0	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
Vegetables (leaf)	64	0.18	0.05	0.71	0.29	3.73
Vegetables (others)	87	0.56	0.05	1.32	4.23	5.40
vegetables (others) raw	50	0.51	0.05	1.41	4.10	5.87
vegetables (others) processed	37	0.63	0,.13	1.18	3.14	5.06
Vegetables (root)	24	0.17	0.05	0.36	1.17	1.38

Vegetables (others):

This food group included 87 samples out of which 26 had been processed (either dried, smoked, or cooked*).* Identification of the respective values was impossible to achieve.

Table 16: BaP occurrence in the food group "Vegetal oils".

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples > 100 ppb
virgin & extra virgin olive oil	575	48	34	11	2	1	0	0
olive oil	188	57	18	7	4	4	2	0
olive pomace oil	60	21	17	170	9	65	9	9
oil in canned food	115	27	6	21	16	4	2	0
Grape seed oil	44	13	4	16	11	2	2	0
sunflower oil	107	20	15	30	28	0	0	1

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
virgin & extra virgin olive oil	671	0.40	0.19	1.39	1.20	3.09
olive oil	280	1.70	0.25	8.12	2.53	33.73
olive pomace oil	268	17.70	9.57	28.30	70.45	134.96
oil in canned food	191	2.62	0.29	7.81	9.60	45.57
Grape seed oil	92	4.20	0.58	10.61	16.86	59.17
sunflower oil	201	3.12	0.40	16.47	9.0	13.0

Sampling on vegetable oils has been targeted in many cases; given a general contamination issue was highlighted in olive pomace oil, the data reflect what had been present on the market. The levels in pomace oil have been subsequently brought down in response to measures and raised awareness to a processing problem which was the cause of the contamination.

Table 17: BaP occurrence in the food group "Waters".

Given Germany is the only MS to have provided data for waters, these should be considered as non representative of exposure of european citizen. However, it seemed relevant to include the data in the report due to the significant amount of knowledge they represent.

option: samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2 (medium bound)

food group	Nb samples 0-0.5 ppb	Nb samples 0.5-1 ppb	Nb samples 1-2 ppb	Nb samples 2-5 ppb	Nb Samples 5-20 ppb	Nb samples 20-50 ppb	Nb samples 50-100 ppb	Nb samples >100 ppb
mineral water	219	0	0	0	0	0	0	0
tap water	945	0	0	0	0	0	0	0

unit in µg/kg wet weight

food group	Nb samples	mean	median	standard deviation	95th percentile	99th percentile
mineral water	219	0.002	0.003	0.001	0.0025	0.0025
tap water	945	0.0004	0.0002	0.0006	0.0015	0.0015

2 CONSUMPTION DATA, ESTIMATES OF FOOD CONSUMPTION.

Each MS participant was asked to complete the Table 3 of annex 7: "Estimate of food consumption for a population group" of the questionnaire using data from his national data.

Food consumption surveys and their accompanying comments, that have been received are briefly described below, they often have different protocols of investigation and therefore are not directly comparable.

Belgium

Consumption data (7-day estimated food record) arise from a study of 1997 of 341 teenagers (14-18 year old) in the city of Ghent carried out by the University of Ghent. In addition, although the consumption data used were detailed, on individual basis, from 7 days records and reasonably recent, the data were not representative for the whole Belgian population (age groups and regions).

Denmark

Intake estimates in the Danish diet are based on dietary intake data collected for adults in the Danish nation-wide food consumption survey 1995 (NFA 1996). The dietary intake data were sampled in three periods covering the whole year to take into account possible seasonal variation in dietary habits. The representative sample of adults included a total of 1837 persons (933 women, 904 men) aged 15-80 years with an average body weight at 72 kg. The Danish nation-wide food consumption survey used a 7-day prospective food record with a pre-coded (semi closed) questionnaire with answering categories for the most commonly eaten foods in the Danish diet. From these data it was possible to represent the total diet by the intake of 205 food items classified into 12 main food groups.

Finland

The main food consumption data used in intake estimations in Finland were derived from the 2002 Dietary Survey of Finnish adults aged 24 – 64 years. The methodology used was a 24-hour recall. The data apply to both males and females, but in the final intake estimation mean consumption values have been used.
Reference
FINDIET 2002 Study. Eds. Männistö S., Ovaskainen M-L, Valsta L. Publications of National Public Health Institute B3/2003.

France

"L'Observatoire des Consommations Alimentaires" (OCA) and the "Centre de REcherche et de DOcumentation de la Consommation" (CREDOC) , have carried out a consumption survey named "Enquête Individuelle Nationale Consommation Alimentaire (**INCA**)", representative of the whole French population. Individuals 7 day food consumptions survey, performed in France in 1998/1999, 3003 consumers of to 2-65 year old (1985 adults more than 15 year old and 1018 infants and children). The methodology used was a pre-coded 7-day record and data refer to food as consumed with weight of food using photographs of foods items.

Germany

The german consumption data arose from the national consumption study (Nationale Verzehrsstudie, NVS) from 1985-88 of 24632 peoples aged ≥ 4 years. The coverage was

national but without the new federal Länders. The methology used was a 7 day dietary records of all consumed food and beverages.

Intake data were calculated for total population and consumers only in the following groups :
Adults (> 18 year old), bw. mean=70.5 kg n=19115
Girls (4-6 year old), bw. mean=20.7 kg n=453
Children (10-12 year old), bw. mean=40.9 kg n=685
Youth (15-17 year old), bw. mean=62.0 kg n=933

Reference for the dietary survey:
Materialien zur Gesundheitsforschung
Schriftenreihe zum Programm der Bundesregierung
Forschung und Entwicklung im Dienste der Gesundheit
Band 18
Die Nationale Verzehrsstudie, Ergebnisse der Basisauswertung. Bonn 1991
ISBN 3-89429-079-X

Greece

The submitted consumption data are average availability (g or mL/person/day) of selected food items among the Greek population. They are based on data retrieved from the DAFNE databank and collected in the context of the Greek national household budget survey, conducted 1998-1999 among 6258 Greek households.

Household budget surveys are periodically undertaken, using nationally representative samples of households. The surveys aim at collecting data on food availability taking into consideration the household purchases, together with contributions from own production and food items offered to members as gifts. The data on food quantities refer to foods available at household level, since no information is recorded on the quantity of foods purchased to be consumed outside the household.

Data were collected year round to allow for seasonal variability in food consumption
Individual availability was estimated without making allowances for edible proportion and under the assumption of equal distribution of food within the household and during the survey period. There are no details about the age or the bodyweight.

Ireland

The North/South Ireland Food Consumption Survey (NSIFCS) investigated habitual food and beverage consumption, lifestyle, health indicators and attitudes to food and health in a representative sample (n=1379) of the 18-64 year old adult population in the Republic of Ireland and Northern Ireland during 1997-1999. The extensive electronic database which has been compiled from this survey is the most complete and up-to-date collection of food consumption data available for adults in the island of Ireland. For the purpose of this SCOOP Task only data for the Republic of Ireland (n=958) was used.

A sample of 1379 adults (ROI n= 958) between the ages of 18 and 64 years, excluding pregnant and lactating mothers, from all over Ireland, took part in the survey. The participants were randomly selected using the electoral register as the sampling frame. Each person who was selected was contacted by mail and followed up shortly afterwards with a visit from a researcher. Eligible persons were invited to participate and a consent form was signed. The response rate was 63%, which was high given the sizeable commitment to the study on the part of the respondents. Analysis of the demographic profile of the sample has confirmed that

the survey sample was representative of the population of the island of Ireland with respect to age, sex, geographical location (urban/rural), marital status, social class and socio-economic group.
Food intake was determined using a 7-day estimated food record. The respondent kept a diary of everything he/she ate and drank over a one-week period, recording the time, location, amount, cooking method and quantity of each item of food and drink consumed. To ensure a consistently high level of detail and accuracy of recording, a researcher visited the respondent (home or workplace) four times during the recording week.
A comprehensive quantification protocol, which included a combination of food quantification methods, was used to obtain the best estimates of food and drink consumed. The researcher weighed some foods e.g. breakfast cereals, spreading fats and beverages. A specially designed photographic food atlas was used to assign weights to other foods. Respondents were encouraged to keep food packaging to allow improved accuracy of the food and portion descriptions.
Self-administered questionnaires were used to collect information on employment status, social and demographic variables, lifestyle factors, habitual physical activity levels, attitudes, health status, medication, supplement use and dieting habits. The researcher carried out body measurements, including height, weight, waist and hip circumference and body composition (i.e. fat/lean ratio using bioeletrical impedance analysis).

The survey commenced in 1997 with a feasibility study to evaluate the proposed methods and to estimate response rates. The main phase of the fieldwork started in October 1997 and data collection was completed in October 1999. Data collection was seasonally balanced and compiled into a fully integrated relational database. This means that each piece of data collected for each respondent is linked to that respondent's ID number.
Quality control procedures were implemented throughout the collection, processing and compilation of data.
The survey was designed to provide data on an all-Ireland basis, however the database is structured so that the data can be analyzed for the island as a whole or separately for the North and South.

Consumption data are available for general population and consumers only.
Average weight: 75 kg
95th percentile weight: 101 kg.

Italy

A food consumption survey conducted in the early 1980's (Turrini et al. 1991) was used to build the reference diet samples (table 18) analysed to estimate the PAH intake (Turrio-Baldassari et al. 1996). It was on a "family" basis: i.e. consumptions were recorded for ca. 10,000 families, then these consumptions were expressed in 'individual' amounts by a system of coefficients which took into account the participation of different members of the family in the overall familiar consumption. Hence, the personal food intakes (table 18 and annex 8) refer to "total population", not to adults.

The most updated Italian data available are from a nationwide survey based on a 7-day food record (Turrini *et al*., 2001). Sampled subjects were 1147 households randomly selected to be representative of four geographical areas, 1978 individuals were analyzed. The results are reported in annex 7.
Intakes of 65 food items are given, as mean ± s.d. and median, for: (a) "total sample" and "consumers only"; (b) males and females; (c) children (1-9 year old), adolescents (10-17), adults (18-64) and elderly (>64); (d) north-west, north-east, centre, south.

Table 18: Mean food intakes (g/person x day) in different Italian areas

		Diets			
			North-	North-	
Food class	Items	National	western	eastern	Southern
Cereals	13	290.5	248.4	262.8	319.7
Meat/Fish	22	177.5	177.8	187.6	164.2
Milk/cheese	9	251.1	254.8	299.6	227.4
Eggs	2	23.6	22.9	22.0	23.0
Legumes	5	26.0	22.4	21.1	28.8
Oils/fats	7	54.3	47.9	56.9	54.9
Fruits/vegetables	36	514.8	451.2	461.1	505.6
Sugar/sweets	14	70.3	81.3	87.3	59.1
Beverages [a]	7	90.1	168.4	149.7	44.3
Wine/beer/spirits	6	177.3	166.7	212.1	159.3
Various/ready	8	10.7	15.3	12.1	7.6
Total	129	1686.2	1657.1	1773.3	1593.9

From: Turrio-Baldassarri *et al*., 1996. These intakes were used to estimate the PAH intakes (annex 8).

[a] Alcohol free, including mineral water, which accounts for most of the difference, in this class, between the southern diet and the others.

Netherlands

Average for both consumers and non-consumers whole population (2-day dietary survey, n=6250, bw = 65.8 kg).
1-6 year olds (2-day dietary survey, n=530, bw = 17.1 kg).

Norway

NORKOST 1997: In the national representative dietary survey NORKOST 1997, 2672 adults aged 16 to 79 participated. The method used in NORKOST was a quantitative food frequency questionnaire, which was distributed and collected in four different periods year round. The result from the survey describes the usual diet among the participants.
5 food items: shellfish, smoked meat, smoked sausage, bacon, oils.
Adults n = 1291, men, age 16-79, average weight =81 kg.
Adults n = 1381, women, age 16-79, average weight = 66kg.

UNGKOST 2000: The survey was carried out in Norway in the autumn of 2000. The sample consisted of children aged 9 and 13 years. The methodology used was a pre-coded 4-day records and data refer to food as consumed with weight of food using photographs of food items.
3 food items: shellfish, smoked sausage, oils.
Children n = 1009, age 13, average weight =49 kg.
Children n = 815, age 9, average weight =32 kg.

Fish & game study: In 1999-2000 two dietary surveys were conducted, focusing on the consumption patterns of foods which may contain environmental contaminants, primarily cadmium, mercury, polychlorinated biphenyls (PCB) and dioxins. Part A of the survey encompassed a nation-wide postal qualitative food frequency questionnaire to 10 000 randomly chosen individuals 18 to 79 years old. Part B encompassed a postal quantitative food frequency questionnaire to 6 000 randomly chosen persons from 14 coastal and 13 inland communities. Standard portion sizes for each of the food items included in the survey were used to convert food frequencies into amounts consumed per day.

Fish & crustaceans
Adults n=2874, men, 18-79 years average weight=82 kg
Adults n=3091, women, 18-79 years average weight=66 kg.

Portugal

Consumption data of vegetable oils. Data retrieved from the National Dietary Survey 1980, according to the information given by Prof. AMORIM CRUZ , from National Institute for Health.
These data are based on a study population n = 9773, all population, age > 15 years old.

Spain

Nature of the survey : Purchase record.
Methodology to collect the data : Basket of the market / daily for 1 year.
Households n = 6000, all population.
Reference of the survey: La Alimentación en España,2000 [Ministerio de Agricultura, Pesca y Alimentación. La Alimentación en España, 2000 (Madrid, 2001). ISBN 84-491-0523-4].

United Kingdom

Mean consumption figures (g/day) are given both by consumer and by population. Standard deviation, median and 97.5th (Note: not 95th) percentile consumption data are given by consumer only. The consumption data are presented for each of the 20 food groups of the Total Diet Study. These food groups are described in Table 2 of each workbook. For each food group the set of recorded consumers will be a different sub-set of the total number of recorded consumers in the dietary survey of interest. The figures for a given food group are derived from a consideration of the food consumption of each individual recorded consumer of that food group.

Adults: The 1986/1987 Dietary and Nutritional Survey of British Adults was a seven day weighed diary study of the diet and nutrition of 2197 adults aged between 16 and 64 chosen to be representative of the general adult population and living in private households. The data have been averaged over 7 days for consumption estimates. Consumption data are generally considered to be representative of an average UK adult but the figures are not targeted towards particular ethnic groups. Pregnant women were not included in the survey because of their different physiology and dietary habits compared to other adults.

Children: The 1997 National Diet and Nutrition Survey (NDNS) of 1701 young people aged 4-18 years was a seven day weighed diary study of the diet and nutrition of schoolchildren chosen to be representative of the general population and living in private households. Consumption data are generally considered to be representative of the general UK young people population but the figures are not targeted towards particular ethnic groups. Consumption data are presented separately for children of the following age ranges: 4-6 years, 7-10 years, 11-14 years and 15-18 years.

Toddlers: The 1992/93 National Diet and Nutrition Survey (NDNS) of children aged 1½-4½ years was a four day weighed diary study of the diet and nutrition of 1,675 children chosen to be representative of the general toddler population and living in private households in Great Britain. It was carried out over 12 months beginning July 1992. Consumption data are generally considered to be representative of an average UK adult but the figures are not targeted towards particular ethnic groups. Consumption data are presented separately for children of the following age ranges: 1½-2½ years, 2½-3½ years and 3½-4½ years.

Consumption data have been submitted for each food group and the following population groups (total population and consumers only):
Adults n = 2197, all population, age 16-64, typical body weight = 60 kg used
Schoolchildren n = 352, all population, age 4-6, individual body weights used
Schoolchildren n = 466 all population, age 7-10, individual body weights used
Schoolchildren n = 443, all population, age 11-14, individual body weights used
Schoolchildren n = 389, all population, age 15-18, individual body weights used
Toddlers n = 576, all population, age 1.5-2.5, 1½-2½ years, individual body weights used
Toddlers n = 606, all population, age 2.5-3.5, 2½-3½ years, individual body weights used
Toddlers n = 493, all population, age 3.5-4.5, 3½-4½ years, individual body weights used

3 PAH INTAKES, ESTIMATED DIETARY EXPOSURES

Participants were asked to complete the questionnaire presented in annex 2. Comments received are briefly described below.
Differences in protocols of investigation made them not directly comparable. PAH intake data are presented, country-by-country in Annex 8.

Because it is the only one for which enough reliable data are available in order to achieve a significant calculation, BaP has been used as an example.

Belgium

An intake estimate was carried out for benzo(a)pyrene.

Only food groups for which at least one sample was above the LOQ of 0.1 µg/kg were taken into account: margarine, sauces, nuts, canned fish, mussels, leafy vegetables, olive oil and similar, other vegetable oil, frying oil, chocolate, chocolate spread.

For these food groups, samples with levels below LOQ were counted as LOQ/2 to calculate mean levels for the food group.

The group of vegetable oils and products was identified as the major contributor to intake. The total average intake of benzo(a)pyrene was calculated to be about 14 ng/person/day or 0.23 ng/kg bw/day.

This is a very raw estimate with a huge uncertainty, because, although the 60 samples covered a broad range of food groups and were taken from unsuspected lots, the number of samples was too limited (very few for each food group, sometimes only one), and only 16 samples contained quantifiable levels of benzo(a)pyrene.

In addition, the quantifiable analytical results have a considerable analytical uncertainty at such low levels.

Denmark

Intake estimates in the Danish diet are based on dietary intake data collected for adults in the Danish nation-wide food consumption survey 1995.

From these data it was possible to represent the total diet by the intake of 205 food items classified into 12 main food groups.

Since Danish data only exists on a few fruit and vegetables and on fish and mussels from oil-polluted areas, the estimation of the intake of benzo(a)pyrene in the Danish diet is supplied with Norwegian data on the concentration of benzo(a)pyrene in foods similar to Danish foods. The contribution of benzo(a)pyrene from rye bread was considered to be important for the Danish diet and therefore, Finnish data on the concentration of benzo(a)pyrene in rye have been included.

The total intake of benzo(a)pyrene from the food groups represented (approximately 30% of the total diet) is 16 ng/person/day (0.2 ng/kg bodyweight/day). Hereof approximately 50% of the intake is from cereals (80% of the Danish intake of cereals is covered by the data given here).

However, benzo(a)pyrene data on wheat flour is missing and is believed to increase the overall contribution of benzo(a)pyrene from the Danish diet.

The intake of smoked mackerel contribute 0.2% to the total diet for an average Danish adult consumer, however the contribution of benzo(a)pyrene are 7% of the estimated intake of benzo(a)pyrene. Sixty five percent (or 15g) of the Danish intake of fish, is due to lack of benzo(a)pyrene data that are not included in the estimate of the intake of benzo(a)pyrene. Food groups such as milk and cheese, raw meat, some fruit and vegetables and beverages such as tap water and wine are not included in the benzo(a)pyrene intake estimate.

For vegetables the few data for carrot and potato covered more than 50% of the total intake per day of this food group. For apples, potatoes and carrots, the concentration of benzo(a)pyrene obtained from these products with 6%, 3% and 14% of the estimated intake of benzo(a)pyrene respectively, correspond to the amount of contribution of the total diet.

For beverages, coffee account for 30% of the total beverage intake, while the concentration of benzo(a)pyrene is so low that only 0.5% of the total estimate of benzo(a)pyrene in the Danish diet comes from coffee.

In a field experiment, vegetables (potatoes, lettuces, carrots and squashs) were grown in two contaminated soils and in a reference soil, whereas fruits were collected from uncontaminated and contaminated private gardens. The results showed elevated levels of PAH in the vegetables from contaminated soil. The contents of contaminants in fruits were generally low and no correlation with the level of contamination in the soils was found. The lettuce and carrot plants were poorly developed in many plots, especially in those containing heavily contaminated soil. The small carrots were irregular and difficult to peel, which may have resulted in remnants of peel in the peeled samples. For all PAH measured, there were no significant differences between concentrations in crops from the two contaminated soils, except for potatoes and carrots without peel. Peeling of the potato and carrot samples reduced the concentrations of benzo(*a*)pyrene and Indeno(1,2,3-*cd*)pyrene considerably. The results of the analyses of PAH in fruits were dominated by results below the detection limit.

For Danish fish data the samples were collected 3 months (June) and 9 months (December) after an oil spill incident in the Danish waters. These data is to be considered as obtained from a polluted area and not suitable for estimation of human consumption. Therefore it is not included on purpose in the estimation of the intake.

Since oil contamination is usually identified by the determination of dibenzothiophene, data on this substance in fish is included.

Finland

In all cases the daily intake was calculated by multiplying the mean occurrence data of each food item or food category by the mean consumption data. In the intake estimation half of all the meat products were thought to be smoked. Calculations were made only in such food groups for which concentration data were available.

In Finland already during the year 1999 a determination of 19 different PAH compounds from a total of 226 systematically selected food samples were made, and the intake of every single PAH compound from foods were estimated. The intake of PAH compounds were most greatly

influenced by meat products and cereals. The total PAH intake was then estimated to be 8.8 microgram/person /day.

The study continued next year and 100 randomly chosen and nationally representative meat and meat products samples were analysed to see the situation and decide, how to get PAH amounts in meat products lowered and checked in in-house-control. Because the results of those hundred meat samples were not satisfied, in addition new 30 targeted samples were taken from those producers, who had problems with high PAH concentrations. This is why at this moment there is so many meat samples, both randomly taken and targeted, compared to other food groups. Another problem is that how many percent of the meat products are smoked is not known. It is estimated that half of the products considered are smoked. Smoking seems to be very popular food process in Finland.

In this SCOOP Task estimation the intake of total PAH compounds is four time higher than earlier estimated. Because more than half of the intake is from meat products it is possible that samples are, however, more targeted than it was meant in the study. The municipal authorities took all the samples, and of course a suspected one will be easily taken.

Because many different PAH-compounds (19) were involved in Finnish intake estimation, the sum of the total PAH is higher than in those countries, where less PAH were involved.

In the calculations, LOQ/2 has been used for the values below the LOQ.

Intake of benzo(*a*)pyrene is 0.27 microgram/person/day. Low molecular PAH compounds dominate and consist of almost the whole intake value.

France

The consumption data are taken from the 1999 INCA survey. This survey was conducted by CREDOC-AFSSA-OCA in 1998-99. It collected data on all the food eaten by the subjects for one whole week. Food consumption data was obtained from consumption diaries, completed for a period of 7 consecutive days. The survey was conducted on 3003 individuals, children and adults, representative of the French population. National reliability was ensured through stratification (age, sex, occupation, social category and size of household). The adult sample comprised 1985 individuals aged 15 and over. Calculations relate only to adult normal reporters, namely 1474 individuals. The children's sample covered 1018 individuals aged between 3 and 14. As there was no available formula for selecting under-reporter individuals, this sample was not corrected.
The food samples were grouped into 14 categories.

Assessment of exposure to PAHs ingested with foodstuffs is based on 978 contamination data as part of targeted or random inspection plans conducted in 1999 and 2001 and surveillance plans conducted between 2000 and 2002 following the wreck of the Erika. The samples concern foods as consumed, with no assumptions as to preparation prior to consumption.

The exposure estimate is based on the hypothesis that a given individual is exposed throughout his lifetime to a mean contamination. Exposure for each individual was calculated by multiplying individual consumption by mean contamination for each food category. This approach enables account to be taken of consumption variability within the population. The result obtained is therefore an exposure distribution.

Exposure was calculated in BaP alone, as a total[3] (sum of the 6 PAHs) and in TEQ (for the 6 PAH) for the series of data with the value for the quantification limit divided by 2 (LOQ/2) allocated to a "not detected" result.

Estimate of the exposure to PAH classified as 2A and 2B by IARC can be considered as being representative of the basic exposure to which the French population is subject via the diet. Although children's consumption is lower than adults, because their ratio of food consumption to body weight is higher than for adults, children are more exposed to the constituents of food.

Due to their high levels of consumption, cereal products contribute 38% of PAH intake in adults and 43% in children. The other dominant source of intake is from meat products (27% in adults and 25% in children). Fish, consumption of which is only one third lower than meat products, has very low contamination levels and so contributes very little (2.4% in adults and 2.2% in children). With the exception of the highly heterogeneous cereal products category, no foods which are specifically vectors for PAH stand out. However, in view of the low number of samples in certain categories and their heterogeneity, the scope of these results is to be understood as limited.

It should also be noted that the results presented do not take account of all the food categories, notably meats and products consumed after cooking (domestic or commercial) which are important potential vectors for PAH for this exposure route. There are few studies available which evaluate the quantity of PAH formed during the cooking of meat, based on the process used (barbecue, grilling, frying, on a hot stone, etc.). Mottier *et al* (2000) showed that mutton sausages cooked on a barbecue contained 14 µg/kg of the sum of 6 PAH. Saint-Aubert *et al* (1992) measured PAH in different samples of meat and fish grilled on two types of gas barbecue, horizontal and vertical. The horizontal barbecue induced the formation of 10 to 30 times more PAH than the vertical barbecue which prevented the over-combustion of fats on the flame of the barbecue (SCF, 2002).

Given that more than 50% of the data corresponds to "not detected" values and that the quantification limits for some PAH are relatively high, exposure was calculated by allocating the value LOQ/2 to the "not detected" results. The exposure estimate gives a result which is approximately twice as high as in the case of the low hypothesis (not detected = 0).

Italy

Italian intakes were estimated for four (three regional and one national) mean diet samples and are reported in Annex 8 (Turrio-Baldassarri et al., 1996). Hence, no intake is available for individual food items or groups.
Food intakes were determined from a previous 10,000 household survey (Turrini et al., 1991) for four geographical areas (north-eastern, west-eastern, southern, whole nation). Food and beverages items included in each reference diet were 129. The consumption survey was conducted in 1980-84.
The reference diet samples were built accordingly, foods being collected in 1993-95. Local food items were purchased in markets of the areas studied; food and beverages were prepared according to the prevalent regional habits.
A 5-day diet sample (about 8.5 kg) was set-up for each diet. The whole sample was then homogenised by means of a laboratory mixer. The market basket composition was based on the mean food intake, as reported in table 18 above.

[3] total: the quantity of PAH measured in the food products.

United Kingdom

All estimated dietary exposures are presented in units of ng/kg bodyweight/day. A standard bodyweight of 60 kg was used for adults. For children and toddlers the individual bodyweights of the recorded consumers were used.

Dietary exposures are presented for each of the 20 food groups of the Total Diet Study plus the estimated dietary exposure from the whole diet.

The total consumer mean, standard deviation, median and high level dietary exposures are derived from the food consumption of all of the recorded consumers in the dietary survey of interest. They are not equal to the sum of the individual exposures from the different food groups, which are all derived from sub-sets of all of the recorded consumers of the whole diet. In all other respects the presentation of the dietary exposure data mirrors that of the food consumption data.

Dietary exposures have not been estimated from the olive-pomace oil, olive oil and extra virgin olive oil samples as those samples were targeted towards potentially contaminated samples and were not therefore representative of oils on the UK market.

4 SUMMARY OF NATIONAL ESTIMATES of BaP *(mean)* INTAKES

Belgium : <u>14 ng/person/day</u>
- ✓ Main contributing food groups : Vegetable oils
- ✓ Samples < LOQ were assigned a value of LOQ/2.

Denmark : 16 ng/person/day
- ✓ Main contributing food groups: bread, potatoes, vegetable oils
- ✓ Samples < LOD were assigned a value of LOD.

Finland : 270 ng/person/day.
- ✓ Main contributing food groups: smoked meat products, bread, cereals
- ✓ Samples < LOQ were assigned a value of LOQ/2.

France : 90 ng/person/day.
- ✓ Main contributing food groups: Bread and cereal products, pizzas and quiches, sausages and ham, fish and smoked fish, shellfish, pastry goods
- ✓ Samples < LOQ were assigned a value of LOQ/2.

Germany : 67 ng/person/day
- ✓ Main contributing food groups: Smoked meat, ham/bacon, vegetable oils, smoked fish
- ✓ Samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2.

Greece : 100 ng/person/day.
- ✓ Main contributing food groups: Vegetable oils
- ✓ Samples < LOD were assigned a value of LOD/2.

Italy : 170 ng/person/day
- ✓ possibly reaching 320 ng/person/day depending on regional diet.

Netherlands : 87.5 ng/person/day.
- ✓ Uses PAH occurrence data from the U.K. 2000 TDS
- ✓ Samples < LOD were assigned a value of LOD/2.

Norway : 17 - 26 ng/person/day (women - men)
- ✓ Main contributing food groups: Hamburger, smoked fishes, smoked sausages, bread
- ✓ Samples < LOD were assigned a value of LOD.

Spain : 36 ng/person/day
- ✓ Main contributing food groups : Mussels, olive oil, sausages & ham
- ✓ Samples < LOD or < LOQ were assigned a value of LOD/2 or LOQ/2

U.K : 94 ng/person/day
- ✓ Main contributing food groups: Beverages (soft drinks, coffee, tea), bread, cereals, milk
- ✓ Samples < LOD were assigned a value of LOD.
- ✓ Year 2000 Total Diet Study.

From this list, it can be derived that the estimated average BaP intakes for an European adult range from 14 to 320 ng/person/day among 11 states that provided data[4].

However due to very fundamental variables and discrepancies among member states such as:

- ✓ the food groups considered,
- ✓ the differences in analytical methodologies and procedures used,
- ✓ the confidence in the expression of data including the fact that LOD and LOQ values have been assigned different values in the calculation,
- ✓ the data on an important source of exposure to PAH (cooked fat meat or fish) have not been included),

this value should be understood as based on recognised, limited science background, not covering the total diet, together with strong analytical inconsistancies and therefore considered a very preliminary rough indication that should be used with considerable caution.

[4] Individuals with a high consumption of food highly contaminated with BaP, may have higher intake, eg: Italy estimates.

DISCUSSION

Apart from the recognised and well known differences in eating habits between member states, many difficulties have been encountered during this SCOOP Task in gathering data.

- ✓ Reliability of the samples. The origin of the samples, whether from random sampling, or from targeted or controlled monitoring affects the consecutive use of data. For example, the results from targeted monitoring may reflect particular contamination issues that were under consideration (e.g. : suspicion control), but may not reflect the level generally to be found in food.
- ✓ Reliability and significance of the data :
  - o huge number of data generated by many different laboratories using different analytical methodologies and procedures (under quality assurance or not),
  - o considerable differences in the expression of results, including a great variation in the meaning of results expressed as: not detected, or < LOD or < LOQ together with a wide range between laboratory for these values.
  - o Impact of the numerous LOD and LOQ data on the final expression
- ✓ Variations in the pattern (number and type) of PAH determined,
- ✓ Variations in the constitution of food groups (limited to the lowest in this report),
- ✓ Variations in the procedures for food consumption survey,
- ✓ Data on an important source of exposure to PAH (cooked fat meat or fish) are not taken into account (such data were too scarce to guarantee reliability and to allow significant considerations).

In order to allow a common European approach for further collection of data and control monitoring, it is highly desirable that EU harmonisation is developed in this area.

1)° Analytical determination.

Analytical determination will require harmonisation at various levels.

✓ *Technical*:

Assay procedures for a given substance in a given matrix may widely vary from one laboratory to another. This is a normal occurrence mainly related to uses, apparatus/device availability, and/or financial limitations. Nevertheless, these factors can broadly influence the reliability of the data and therefore the capacity to compare data from one MS to another.

The many possible confounding factors include:

- Sampling and treatment of the samples: origin, handling, storage conditions, processing (extraction, separation and clean up).
- Analytical methodologies and procedures used, variability in the limits of detection and of quantification even when using the same methodology and procedure, uncertainty, quality assurance information.
- Expression of results wet/dry weight, reference. How to make use of the data expressed as "not identified" or "not quantifiable", given the wide inter-laboratory variations in the meaning (and estimated level) of these definitions.

✓ *Administrative*:

Conditions of generation and collection of data may vary, from one MS to another or within a single MS. The *complete, consistent and comparable* expression of data can be questionable

and may depend on the context, *e.g*. data generated in the case of a survey or control monitoring can be expressed merely as above or under the respective limit value, studies performed by research laboratories may follow different procedures than those carried out in administrative laboratories.

2°) Selection of PAH.

Selection of PAH to be measured should be recommended, taking into account toxicological properties as well as the pathways of occurrence which largely influence the PAH patterns.

- high temperature grilling or barbecuing, particularly on an open flame.
- use of non suitable (type of wood or charcoal.) or inadequate combustible (plastic, tyres..)
- high temperature food processing procedures.
- drying processes (oleaginous seeds, cereals, …).
- certain food smoking processes.

Regarding hot smoking processes and drying processes for cereals, fruit, spices and other products such as fish products, new and more targeted contamination data need to be collected to enable the determination of contamination profiles and the impact of various cooking and processing parameters.

Exposure from consumption of water has not been addressed in this report because both PAH contamination is likely to be very low and the available data were from one single country (Germany). This issue deserves however consideration and needs assessment.

CONCLUSIONS & RECOMMENDATIONS

Various foods gathered into 44 food groups have been analysed in the 14 participating countries (13 Member States plus Norway) for the presence of PAH. The only consistently tested PAH (in 99% of the sample) was benzo(a)pyrene. Thirty other PAH were tested for, to varying extents, but not enough to be able to establish clear patterns or relationships between the levels of the different PAH. As a first outcome, the distribution is unbalanced given that 5 food groups gathered more than 80% of the samples (total number of samples = 8861) : Sausages & ham (2358 samples, 27%), Vegetal oils (2110 samples, 24%), Fish / fish products (1173 samples, 13%), Waters (excluding tap water) (1004 samples, 11%), Meat (519 samples, 6%). It should be noticed that almost no data were available regarding home cooked (e.g. barbecued, fried, roasted) food.

Highest levels for BaP were found in: dried fruits (48.10 µg/kg wet weight), olive pomace oil (17.7µg/kg), smoked fish (5.28µg/kg), grape seed oil (4.2µg/kg), smoked meat products (3.27µg/kg), fresh molluscs (3.09µg/kg), spices/sauces and condiments (2.16µg/kg).

In view of the different approaches to collect data in the participating States and the differing amounts of available information to accompany the data, it is difficult to draw firm conclusions. In particular, it is often difficult to differentiate between those samples which have been targeted at known food sources of contamination and those samples which have been randomly taken for foods representative of what is available on the market.

From the available data, the estimated average BaP intakes for an European adult range from 14 to 320 ng/person/day among the 11 states that provided intakes data. These values are merely indicative, they have very limited significance and do not represent a reliable assessment.

FUTURE NEEDS.

It should be underlined that the participating countries have expressed great interest in the SCOOP Task by a recognised and voluntary commitment in both the meeting discussions that took place during implementation of the Task and involvement in the communication of numerous data. This is further illustrated by the fact that in the course of the SCOOP Task, some countries have even carried out new determinations on topics identified as worth needing more data.

It is noticeable that most of the determinations have been carried out following episodes of contamination for a given food. Therefore, there is a lot of data on those foods that are the most likely to be affected by these episodes. This partly contributes to an erroneous, or at least impaired evaluation of the "usual" contamination of the diet by PAH. But in line with one of this SCOOP Task requirements, allowed to spot which foods may be the most likely targets of high contamination.

Harmonisation for the collection of data appears as an utmost priority In order to allow a common European approach for further improved collection of data and control monitoring, it is highly desirable that such a harmonisation and improvement starts as soon as possible and is handled at the level of the European Union.

In order to ensure reliable and comparable information, it is highly advisable to further develop inter-laboratory trials.