JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

J. Agric. Food Chem. **2008,** *56,* 6039–6045 **6039**

Dietary Acrylamide Exposure Estimates for the United Kingdom and Ireland: Comparison between Semiprobabilistic and Probabilistic Exposure Models

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Since the discovery of acrylamide in foods, there have been many calculations of dietary exposure. Total diet studies have been commonly used to estimate consumer exposure of acrylamide; however, these often fall short in evaluating true exposure levels because of limitations in small occurrence data sets. Dietary exposure to acrylamide can also be estimated by use of modeling packages. The U.K. Food Standards Agency and the Food Safety Authority of Ireland have prepared estimates for dietary acrylamide exposure using semiprobabilistic and probabilistic modeling. Occurrence data were obtained from the European Union acrylamide monitoring database, whereas consumption data were obtained from the relevant U.K. and Irish National Diet and Nutrition Surveys. The mean adult U.K. consumer exposure was estimated as 0.61 μg/kg of body weight (bw)/day and high-level adult consumer exposure (P97.5) as 1.29 μg/kg of bw/day. The mean adult Irish consumer exposure was estimated as 0.59 μg/kg of bw/day and the high-level adult consumer exposure (P97.5) as 1.75 μg/kg of bw/day. Owing to the wide range of acrylamide levels in foods, semiprobabilistic modeling does not always provide an accurate picture of dietary exposure levels and patterns. Therefore, a comparison of semiprobabilistic assessments to probabilistic assessments of U.K. and Irish dietary exposure estimates of certain food groups is provided.

KEYWORDS: Acrylamide; diet; modeling package; semiprobabilistic; probabilistic

INTRODUCTION

Acrylamide is a reactive unsaturated amide that has found industrial uses in the manufacture of polyacrylamides that are used in water treatment, mining, grouting agents, and cosmetics. In 2002, Swedish scientists and the Swedish National Food Authority reported the discovery of acrylamide in a variety of fried and baked foods (*1, 2*). Other researchers soon verified the Swedish findings, reporting up to milligrams per kilogram quantities of acrylamide in carbohydrate-rich foods that had been subjected to high-temperature cooking/processing (*3–6*). Acrylamide has been shown to be neurotoxic in humans (*7–9*) and has been shown to induce tumors in laboratory rats (*10, 11*); it has also been classified as a probable human carcinogen (*12*), and as such several international bodies have concluded that dietary exposure should be as low as reasonably achieveable (*13, 14*).

The most significant pathway of formation of acrylamide in foods has been shown to arise from the reaction of reducing sugars with asparagines via the Maillard reaction at temperatures above ca. 120 °C (*15*). Acrylamide has been found in a wide range of heat-treated foods; it is found in both foods processed by manufacturers and foods that are cooked in the home. Several large databases of acrylamide occurrence data have been compiled. These include the European Union's acrylamide monitoring database (*16*), the U.S. Food and Drug Administration's acrylamide survey data (*17*), and the World Health Organization's Summary Information and Global Health Trends database for acrylamide (*18*). All of these databases show that acrylamide is most prevalent in fried potato products (such as French fries and potato chips), cereals, bakery wares, and coffee. Because many of these foods represent dietary staples in the Western world, there has been much interest in dietary exposure to acrylamide.

There have been many estimates of dietary exposure to acrylamide. In, 2002 the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) reviewed the existing data on exposure and concluded that long-term acrylamide exposures would be in the range of 0.3−0.8 μg/kg of body weight (bw)/day (*14*). The committee stressed that data were sparse and further work should be undertaken to produce more robust exposure assessments taking into account all dietary sources of acrylamide. Similar recommendations were also given by the European Commission's Scientific Committee on Food (*13*).

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10.1021/jf073050x CCC: $40.75
Published on Web 07/15/2008

Table 1. Acrylamide Levels in Foods Reported in the Literature

food group	food subgroup	no. of samples	acrylamide (μg/kg) min	max	mean[a]
bakery wares	white bread	7	60	117	84
	wholemeal bread	6	15	33	17
	rye bread	38	10	397	140
	other bread	9	15	60	24
	crackers excluding sweet crackers	43	10	830	301
	other ordinary bakery products	32	10	1987	137
	crisp bread and crisp rolls	212	10	2380	411
	bread type products	29	10	514	140
	cakes, cookies, and pies	135	10	1080	202
	other fine bakery products	5	169	1491	557
beverages (coffee and beer)	coffee (as reported)	273	80	2932	600
	beer and lager	37	10	10	5
	cider	1	10	10	5
	malt drinks	3	90	130	107
biscuits	miscellaneous biscuits	189	10	1950	303
	gingerbread	680	10	7834	569
	ginger biscuits	139	15	2220	585
	almond based	79	10	1234	310
	shortbread	151	10	6798	409
cereals and cereal products	whole, broken, or flaked grain	4	10	30	15
	flours and starch	3	13	112	42
	muesli	51	10	258	64
	maize-based cereals	110	10	545	98
	rice-based cereals	11	20	1649	251
	wheat-based cereals	22	30	532	132
	mixed grain cereals	15	50	260	137
	oat-based cereals	6	10	274	95
	bran-based	15	20	640	304
	rice cakes	8	15	250	137
confectionery	chocolate confectionary	47	10	826	138
	sugar based confectionary	23	10	548	151
fruits, vegetables, and nuts	dried fruit	73	10	258	42
	fruit in vinegar, oil, or brine	32	3	1548	169
	fresh vegetables	5	15	20	13
	dried vegetables	3	220	439	303
	canned vegetables	27	10	68	10
	nuts and seeds	1	200	200	200
potatoes and potato products	potato chips	349	10	4215	626
	re-formed potato snack products	22	50	1680	818
	french fries	723	10	3428	299
	fried potato products and roast potatoes	35	5	1428	320
	miscellaneous potatoes	1	66	66	66
infant foods	rusks	215	10	1060	143
snack products	maize-based snacks	42	40	820	201
	pretzels	10	60	273	165
sugar syrup	sugar-based syrups	3	15	438	156

[a] Medium bound.

In this study an indirect approach to estimating dietary exposure was followed by combining an independently gathered data set of acrylamide occurrence with existing food consumption information from both the United Kingdom and Ireland.

Three different calculation methods can be employed when using indirect exposure models. In order of increasing complexity, these are point estimates (sometimes referred to as deterministic methods), semiprobabilistic methods, and probabilistic methods. Point estimates involve combination of one contaminant level (single or statistically derived value) and one consumption level (usually statistically derived value) for each food. These are then summed over all foods to estimate an overall contaminant dietary exposure. The result is one level of exposure that does not provide any information on how many people have such an exposure or how many exceed the calculated exposure. The semiprobabilistic approach combines one contaminant level (usually the mean or median) with a distribution of consumption levels, resulting in a distribution of exposure defined by the variation in consumption levels. In probabilistic modeling, a distribution of contaminant levels is combined with a distribution of consumption levels, resulting in a distribution of exposures that is generally considered more representative of the true exposure.

The previous U.K. estimate of acrylamide dietary exposure was published in 2005. This estimate was produced using the total diet study approach (*19*), one of four commonly used

Table 2. Acrylamide Exposure Estimates

organization, country	population/sex (age)	estimated dietary intake (μg/kg of bw/day) mean	P95; *, P90; †, P97.5
Bundesinstitut für Risikobewertung (BfR), Germany (*26*)	all (15−18)	1.1	3.2
Norwegian Food Safety Authority (NFAS), Norway (*27*)	males	0.49	1.04*
	females	0.46	0.86*
	males (13)	0.52	1.35*
	females (13)	0.49	1.2*
Agence Française de Securite Sanitaire de Aliments (AFSSA), France (*28*)	all (>15)	0.5	0.98
	all (3−14)	1.25	2.54
Livsmedelsverket (SNFA), Sweden (*29*)	all (18−74)	0.45	1.03
Food and Consumer Product Safety Authority (VWA), The Netherlands (*30*)	all (1−97)	0.48	0.60
	all (1−6)	1.04	1.1
	all (7−18)	0.71	0.9
Food and Drug Administration (FDA), United States (*31*)	all (2+)	0.44	0.95*
	all (2−5)	1.06	2.33*
Food Standards Agency (FSA), United Kingdom[a]	all (19−64)	0.61	1.29†
Food Safety Authority of Ireland (FSAI), Republic of Ireland[a]	all (18−64)	0.59	1.75†

[a] Consumer estimates (this study).

Table 3. U.K. Dietary Exposure Estimates (Semiprobabilistic Determination)

food group	dietary exposure to acrylamide (μg/kg of bw/day) mean consumer	high-level consumer (P97.5)	total population mean
bakery wares	0.15	0.43	0.15
beverages	0.04	0.16	0.03
biscuits	0.05	0.21	0.03
cereals	0.08	0.23	0.08
confectionery	0.02	0.10	0.02
fruit, vegetables, and nuts	0.02	0.07	0.02
infant foods (rusks)	0.01	0.02	0.00
potatoes and potato products	0.27	0.85	0.22
snack products	0.02	0.08	0.00
sugar-based syrups	0.01	0.03	0.00
total (all food groups)	0.61	1.29	0.56

methods to estimate dietary exposure. The other three are duplicate diet studies, total, hypothetical/simulated diet methods, and methods combining estimates from food consumption and monitoring programs; details of these methods can be found elsewhere (*20*).

Exposures of 0.3 and 0.6 μg/kg of bw/day were calculated for mean and high-level adult consumers, respectively. These values are at the low end of the range of dietary acrylamide exposure considered by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) during their 2005 review of acrylamide (*21*). Evaluation of the U.K.'s 2005 estimate reveals several limitations: food samples were composited prior to analysis; thus, instances of high acrylamide contamination were diluted; the 2003 potato samples contained no fried potato products or crisps (samples that are expected to contain high levels of acrylamide); the miscellaneous cereals samples were dominated by pasta, and there were no samples of crisp bread (another food that contains high levels of acrylamide).

Table 4. Irish Dietary Exposure Estimates (Probabilistic Determination)

food group	dietary exposure to acrylamide (μg/kg of bw/day) mean consumer	high-level consumer (P97.5)	total population mean
biscuits	0.08	0.34	0.06
bread	0.20	0.72	0.20
breakfast cereals	0.05	0.18	0.03
cocoa products	0.01	0.08	0.00
coffee	0.02	0.09	0.01
potatoes and potato products	0.33	1.36	0.29
total (all food groups)	0.59	1.75	0.59

MATERIALS AND METHODS

The aims of the current work were to update the U.K. acrylamide exposure estimate and to compare this estimate to Irish estimates of dietary exposure from certain food groups. To compare these estimates, the same occurrence data sets and approaches for calculating dietary exposure were adopted. Because the U.K. Food Standards Agency uses a semiprobabilistic modeling package and the Food Safety Authority

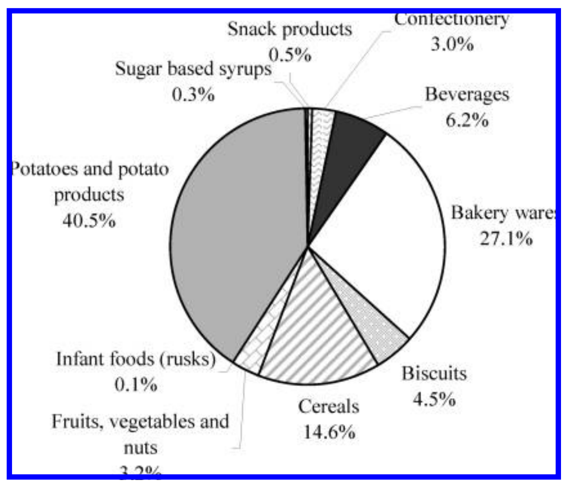

Figure 1. Contribution to acrylamide intake from all food groups for the U.K. adult population.

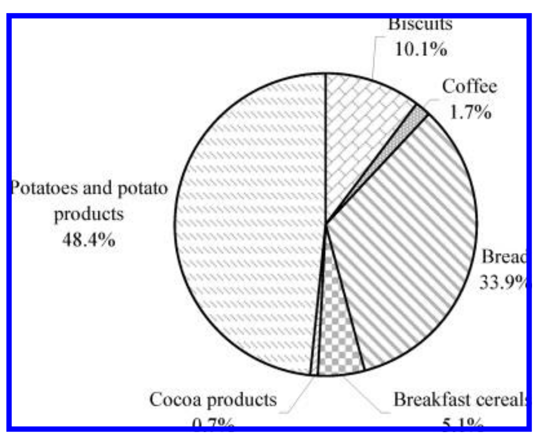

Figure 2. Contribution to acrylamide intake from selected food groups for the Irish adult population.

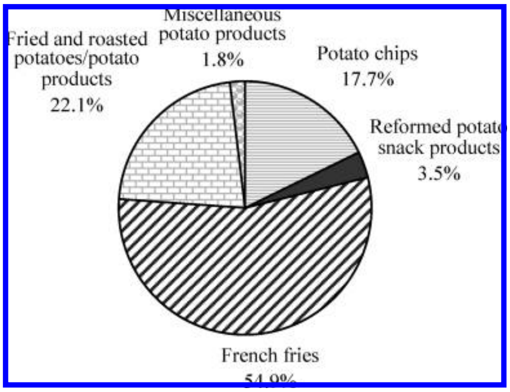

Figure 3. Contributions to acrylamide intake for the U.K. adult population from potatoes and potato products.

of Ireland uses a probabilistic model, it was necessary to compare these two models before being able to make comparisons between the U.K. and Irish assessments.

Occurrence data were obtained from the European Union (EU) acrylamide monitoring database (*16*). This database contains data on levels of acrylamide in over 7000 samples of foods collected by EU member states and the European food industry. The occurrence data have been collected since 2002 and are subject to eight exclusion criteria to ensure robustness of the database. Additional data were also obtained from the Dublin Public Analyst's Laboratory. To combine the occurrence database with the relevant consumption databases, it was necessary to group the individual samples into food groups. This grouping of foods was achieved using the Codex classification system, with some modifications to allow for the nature of acrylamide occurrence in foods. **Table 1** lists the food groups (and their subgroups) that were used in this study, along with the mean, minimum, and maximum levels of acrylamide found in those groups. A considerable number of entries in the EU acrylamide monitoring database were excluded from this study owing to insufficient description of the exact type of food/beverage. All occurrence data used in this study were medium bound, that is to say, that when levels of acrylamide were reported as being less than the limit of detection for the analytical method used for acrylamide determination, the level of acrylamide was taken to be equal to half the value of the limit of detection.

When ratifying the data from the EU acrylamide monitoring database, we assumed that all acrylamide levels reported for coffee and drinking chocolate were based on analysis of the raw material; many entries in the database did not make clear whether the coffee/drinking chocolate was analyzed as sold (raw product) or as ready to consume (thus diluted). This assumption was made on the basis that reported levels of acrylamide in coffee and drinking chocolate compared well to those reported by the Dublin Public Analyst's Laboratory for coffee and drinking chocolate as sold (raw product). A dilution factor of 100 was used to combine data on acrylamide levels in these beverages with consumption data. The dilution factor equates to 2.5 g of instant coffee in 250 mL of water to represent one heaping teaspoon of coffee per cup (*22*). This assumption is in line with manufacturer's recommendations.

Consumption data were obtained from the relevant national surveys. U.K. consumption data were taken from the National Diet and Nutrition Survey for adults (2000). This survey aimed to provide a comprehensive assessment of the dietary habits and nutritional status of the U.K. population. Two thousand adults were surveyed, although only 1724 respondents provided enough information to be included in this study. Irish consumption data were obtained from the North/South Ireland Food Consumption Survey (Adults 1997−1999). This survey 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. For the purpose of calculating dietary acrylamide exposure, only data for the Republic of Ireland (n = 958) were used. Both consumption surveys were conducted over 7 days and involved the keeping of a food diary detailing each consumption event; these surveys have been reported elsewhere (*23, 24*).

U.K. dietary exposure estimates were made using the Food Standards Agency's semiprobabilistic in-house intake model. This model is a custom-made statistical program that can combine individual dietary survey records taken from the National Diet and Nutrition Surveys with single values of a chemical concentration in food. Chemical concentrations can be entered for each food subgroup being considered, and the program will combine these data with each participant's food diary in the relevant consumption survey. When a particular food is eaten, consumption is combined with the relevant chemical concentration for each participant in the survey from all of the specified foods. All exposures were calculated as chronic (each participant's average daily exposure over the length of the survey, 7 days, was calculated). The full distribution of participants' exposure is then calculated, and from this distribution exposure statistics are extracted. Dietary exposure estimates for the Republic of Ireland were made using the probabilistic CREMe 2.0 Food model (CREMe Software Ltd., O'Reilly Institute, Trinity College, Dublin, Ireland). This program is a unique tool that uses high-performance computing to allow accurate estimate of exposure to contaminants, food additives, food packaging migratory compounds, novel foods, nutrients, pesticide residues, and microbial contaminants. The main input components are contaminant concentration data, food consumption data, bioavailability data, food processing data and the effect on foods and chemicals, recipe and food grouping data, and market share/brand loyalty data. These data sets are combined using the CREMe model to allow accurate and efficient exposure assessments. In the current probabilistic assessments, distributions of acrylamide occurrence and consumer consumption were combined via Monte Carlo type calculations to yield distributions of dietary exposure.

In addition, U.K. dietary acrylamide exposure from potatoes and potato products and biscuits was estimated using the CREMe software to allow for direct comparison of Irish and U.K. exposure estimates.

RESULTS AND DISCUSSION

U.K. Semiprobabilistic Estimate. Mean dietary exposures to acrylamide were calculated as 0.56 μg/kg of bw/day for the total U.K. adult population and at 0.61 μg/kg of bw/day for adult consumers (estimate restricted to only those people in the total population that consume foods containing acrylamide). A total dietary exposure for high-level consumers (P97.5) was estimated at 1.29 μg/kg of bw/day. These estimates are greater than those estimated previously for the U.K. population (see earlier) and compare well to exposure estimates for other populations reported in the literature (see **Table 2**). These new U.K. estimates also compare well with those reported by JECFA, which ranged from 0.3 to 2.0 μg/kg of bw/day for mean consumers and from 0.6 to 3.5 μg/kg of bw/day for high-level consumers. Direct comparison of the U.K. estimates with those reported in **Table 2** is not possible owing to a number of factors. When comparisons between different exposure estimates are made, it is important to consider the type of estimate (population statistics or consumer statistics) as well as the age range for the population under consideration. The U.K. exposure estimate is for adults in the age range of 19−64. Both population and consumer statistics (see earlier) are presented for U.K. adults, whereas most of the estimates in **Table 2** refer to population statistics only. Several of the estimates given in **Table 2** are for populations of all age ranges, whereas others are for much smaller age ranges. Differences will also arise because of different calculation methods and food categorization. However, our estimates are within the range of these previously reported values.

Irish Probabilistic Estimate. Mean dietary exposures to acrylamide were calculated as 0.59 μg/kg of bw/day for the total Irish adult population and as 0.59 μg/kg of bw/day for adult consumers. A total dietary exposure for high-level consumers (P97.5) was estimated at 1.75 μg/kg of bw/day. It must be noted that these estimates do not take into account all possible dietary sources of acrylamide, only those food groups that are known to be most important. The agreement of the

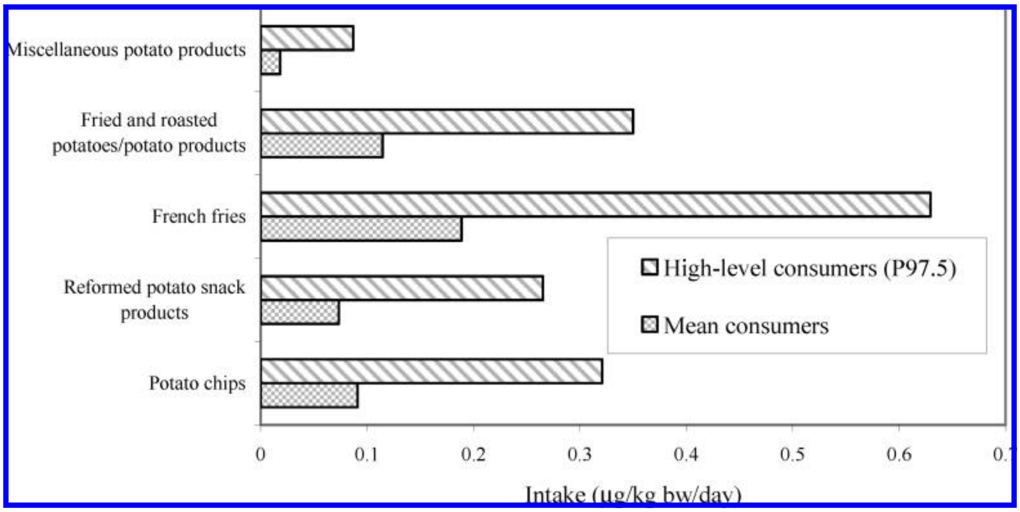

Figure 4. Contributions to acrylamide intake for U.K. adult consumers from potatoes and potato products.

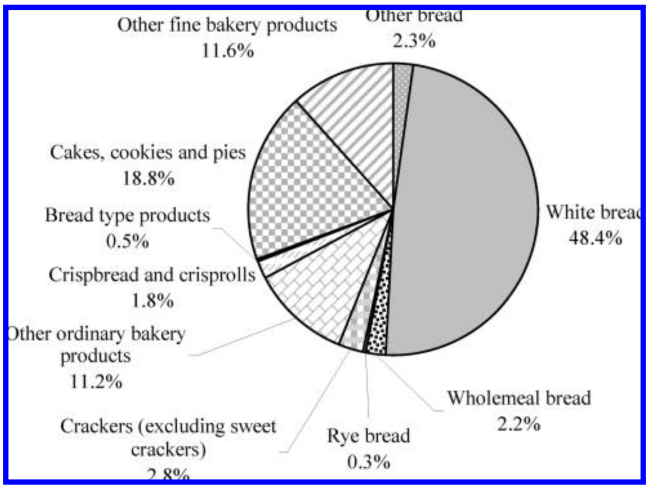

Figure 5. Contributions to acrylamide intake for U.K. adult population from bakery wares.

population and consumer estimates serves to show that acrylamide contamination of food is prevalent in those foods commonly consumed by all Irish adults.

Contributions to total estimated dietary exposure of acrylamide for the United Kingdom and the Republic of Ireland can be found in **Tables 3** and **4**, respectively; note that consumption of infant rusks was found to contribute to estimated adult dietary acrylamide exposure and is thus included in the presented data.

Figure 1 shows the contributions of the different food groups to overall estimated acrylamide exposure for the U.K. adult population, whereas **Figure 2** shows the contributions of selected food groups to overall estimated acrylamide exposure for the Irish adult population. For both U.K. and Irish adults, consumption of potatoes and potato products gives rise to the largest contribution to dietary exposure of acrylamide. Just over one-fourth of the U.K. estimated dietary acrylamide exposure is derived from the consumption of bakery products, whereas almost 15% is the through consumption of cereals and cereal-based products. A similar pattern is observed for the Irish estimated dietary acrylamide exposure, with one-third derived from the consumption of bread and 5% being derived from the consumption of breakfast cereals.

Closer inspection of the U.K. estimates for total population exposure to acrylamide (see **Figure 3**) reveals that of potatoes and potato products, over half the estimated dietary acrylamide exposure is attributable to French fries and just under one-fourth

Table 5. Comparison of U.K. Semiprobabilistic and Probabilistic Estimates of Acrylamide Intake

		acrylamide intake (µg/kg of bw/day)		
method of determination	food group	mean consumer	high-level consumer (P97.5)	total population mean
semiprobabilistic	potatoes and potato products	0.27	0.85	0.23
probabilistic	potatoes and potato products	0.28	1.04	0.25
semiprobabilistic	biscuits	0.05	0.21	0.03
probabilistic	biscuits	0.05	0.25	0.03

Table 6. Comparison of Irish Semiprobabilistic and Probabilistic Estimates of Acrylamide Intake

		acrylamide intake (µg/kg of bw/day)		
method of determination	food group	mean consumer	high-level consumer (P97.5)	total population mean
semiprobabilistic	potatoes and potato products	0.33	1.11	0.28
probabilistic	potatoes and potato products	0.33	1.37	0.29
semiprobabilistic	biscuits	0.08	0.28	0.06
probabilistic	biscuits	0.08	0.34	0.06

is attributable to the consumption of fried and roasted potato products (excluding potato chips). A similar exposure pattern is observed for U.K. consumers (see **Figure 4**); it is interesting to note that for high-level consumers (P97.5), the exposure pattern is also very similar. These similarities are to be expected because potatoes and potato products are a staple of the U.K. diet. **Figure 5**, which displays the relative dietary contributions of the food subgroups that make up the bakery wares group, shows that white bread is a major contributor to dietary acrylamide exposure. This observation is not surprising owing to the relatively high consumption of this food by the U.K. population.

U.K. and Irish Probabilistic Estimates: Direct Comparison of Certain Food Groups. Probabilistic estimates of dietary acrylamide exposure from the consumption of potatoes and potato products and biscuits are compared to semiprobabilistic estimates in **Tables 5** and **6**. Dietary exposure estimates calculated by the two methods are similar; however, in most cases (and for all high-level consumers) U.K. dietary exposures estimated by the probabilistic model are higher than those estimated by the semiprobabilistic model. This same observation is partly apparent in the Irish estimates that were made using the CREMe software in both probabilistic and semiprobabilistic (using a similar method to the U.K. semiprobabilistic calculations) modes. The same occurrence and consumption data have been used in both estimates; thus, the U.K. estimates provide a comparison between the semiprobabilistic dietary exposure model and the probabilistic CREMe model. Meanwhile, the Irish estimates provide a truer comparison between semiprobabilistic and probabilistic estimates owing to the same modeling package being used for both calculations. The higher dietary exposure estimated by the use of probabilistic modeling may be a result of the distributions of occurrence data for both food groups: distributions were log-normal with significant numbers of samples having very high concentrations of acrylamide. Probabilistic modeling takes into account consumption of these high-level acrylamide containing foods, whereas in semiprobabilistic modeling these occurrences of high levels are diluted during calculation of the mean acrylamide levels of food groups. Indeed, the difference between probabilistic and semiprobabilistic dietary exposure estimates is most evident for high-level consumers (see **Tables 5** and **6**). Certainly, this finding requires more investigation as it is possible that the U.K. semiprobabilistic determination for acrylamide exposure may be underestimating for high-level consumers.

Estimated dietary acrylamide exposure through the consumption of potatoes and potato products and biscuits is fairly similar for both the U.K. and Irish total population and consumers, with Irish estimates being slightly higher. The overall U.K. and Irish exposure estimates yield margins of exposure (based on a no observed adverse effect level of 0.2 mg/kg of bw/day) (*25*) that are >110 for morphological nerve changes. These are within the range estimated by JECFA and support the Committee's conclusion that morphological changes in nerves cannot be excluded for some individuals with very high dietary exposure (*21*).

Our findings highlight the need to continue to monitor dietary acrylamide exposure in the U.K. and Irish populations and further examine differences in exposure estimates observed through the employment of different exposure models. In particular, comparison of dietary exposure estimates is compounded by the different groupings of food that are employed by different researchers. Harmonizing dietary exposure assessment methodology will greatly help exposure assessors compare assessments and draw useful conclusions.

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Received for review October 16, 2007. Revised manuscript received January 18, 2008. Accepted May 20, 2008.

JF073050X