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TOTAL AND INORGANIC ARSENIC IN THE 1999 TOTAL DIET STUDY

The Food Standards Agency has completed a survey of total and inorganic arsenic in the 1999 Total Diet Study (TDS). Arsenic is present in food in different chemical forms, with inorganic forms being the most toxic. Most arsenic in the diet is present in the less toxic, organic form. However, it is difficult to reliably measure the forms of arsenic present, so most surveys measure total arsenic.

This is the first survey in which both inorganic and total arsenic concentrations have been measured in the Total Diet Study, which provides information on dietary exposures of the general UK population to chemicals such as nutrients and contaminants. These results have been used to estimate dietary exposures to inorganic and total arsenic for the average UK consumer and to help to identify any foods that make a significant contribution to total exposures to inorganic arsenic.

The key facts of this survey are:

- Concentrations of total arsenic are reported for each of the 20 food groups of the TDS. The concentrations found are generally similar to those reported in previous surveys. The concentration of total arsenic in the fish group has decreased since the last TDS. Fish is the main source of arsenic in the UK diet.

- For the first time, inorganic arsenic is reported for food groups (the miscellaneous cereals group and poultry and fish groups) where the concentration of total arsenic was high enough to allow the measurement of inorganic arsenic. The level of inorganic arsenic in the fish group was low, and less than 1 per cent of that reported for total arsenic.

- Dietary exposures to total and inorganic arsenic for the general UK population and for mean and high level consumers were estimated. For those exposures which have been estimated previously (adult consumer and population exposure to total arsenic), exposures have decreased.

- The independent Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) considered that the results offer reassurance that levels of arsenic in food are not increasing. It concluded that the dietary exposure to arsenic identified in the survey was unlikely to be a concern for consumer health.

Background

Arsenic in Food

Metals and other elements in food are of interest because of their possible health effects. Some, like inorganic arsenic, have no known beneficial biological function and long-term, high-level exposures may be harmful to health (inorganic arsenic is a human carcinogen).

Environmental sources are the main contributors to contamination of food with metals. Arsenic can enter the environment from natural sources, such as rocks and sediments, and as a result of human activities such as coal burning, copper smelting and the processing of mineral ores. Levels of arsenic are higher in the aquatic environment than on land and arsenic is known to accumulate in fish and some shellfish, such as crabs and whelks.

Fish contain relatively high levels of arsenic compared with other foods and are the most significant source of arsenic in the UK diet. In the 1997 UK Total Diet Study,[1] the fish group contained the highest average concentrations of arsenic (4.4 milligram/kilogram) and contributed 94 per cent of the average population dietary exposure to arsenic. In comparison, other food groups from the 1997 TDS survey contained an average arsenic concentration ranging between 0.0004 and 0.007 milligram/kilogram, and each only contributed up to 2 per cent of the average population dietary exposure to arsenic.

Arsenic is present in food in different forms (chemical species) which vary in toxicity, with inorganic forms being the most toxic. Most of the arsenic in the diet is present in the organic forms[3]. Inorganic arsenic species are typically found in food at a level of 1 to 3 per cent. Fish and seafood, which are the most significant sources of arsenic in the diet, predominantly contain organic arsenic species. The main organic arsenic species in fish (more than 90 per cent of total arsenic) is in the form of arsenobetaine which is also the

main form found in crustaceans and bi-valve molluscs.[4]

However, it is difficult to distinguish analytically between the different forms of arsenic in food, and for this reason previous FSA surveys have measured total arsenic.

The Total Diet Study (TDS)

The TDS is an important part of the Food Standards Agency's surveillance programme for chemicals in food and has been carried out on a continuous annual basis since 1966. Foods representing the average UK diet are purchased, prepared and combined into food groups. Results from the TDS are used to estimate dietary exposures of the general UK population to chemicals in food, such as nutrients and contaminants, to identify trends in exposure and make assessments on the safety and nutritional quality of food.

The design of the UK Total Diet Study has been described in detail elsewhere, but in summary involves 119 categories of foods combined into 20 groups of similar foods for analysis.[5] The relative proportion of each food category within a group reflects its importance in the average UK household diet and is based on an average of three previous years of consumption data from the National Food Survey.[6-8] Foods are grouped so that those that may make a significant contribution to dietary exposure (that is, foods known to be susceptible to contamination such as offal and fish, and foods which are consumed in large quantities such as bread, potatoes and milk) are kept separate.

The foods making up the 20 groups are obtained from retail outlets in 24 towns throughout the UK at fortnightly intervals and then transported to one centre where they are prepared.

Each food group obtained from each location (i.e. a total of 480 samples) in the 1999 TDS was analysed for total arsenic, and for those food groups (miscellaneous cereals group and poultry and fish groups) where the concentration of total arsenic was high enough to allow the measurement of inorganic arsenic, this form was also measured. The mean (average) concentrations from the 24 samples of each food group were used together with data on the consumption of these food groups to estimate dietary exposure for the average UK population and for individual consumers. Population exposure estimates are compared with those from previous years to identify trends in exposure.

Brand names

Brand names are not available as TDS samples are composites of a number of different foods.

Methodology

Sample preparation

Individual components of the TDS food groups were purchased from retail outlets in 24 representative towns in the UK in 1999 and prepared as for consumption, including cooking where appropriate, before being combined into one of 20 food groups. Samples were then thoroughly homogenised and frozen until analysis at –20 °C. The 1999 TDS samples were prepared by the Laboratory of the Government Chemist.

Analysis

Samples were analysed in duplicate for total and inorganic arsenic at the Central Science Laboratory (CSL). Total arsenic was measured using both direct nebulisation inductively coupled plasma-mass spectrometry (ICP-MS) and hydride generation ICP-MS for total arsenic. Inorganic arsenic was measured using high resolution ICP-MS.

Total Arsenic

Samples were homogenised and digested in nitric acid by microwave heating. All samples, with the exception of the fish group, were measured using both direct nebulisation ICP-MS and hydride generation ICP-MS, to ensure the best detection limits and accuracy of results. Fish analysis was limited to direct nebulisation ICP-MS as an earlier survey[1] had shown that this is a more appropriate technique for measuring total arsenic in fish as the largely organic species of arsenic in fish are not detected by hydride generation -ICP-MS.

Inorganic Arsenic

Samples were dissolved with hydrochloric acid and extracted into chloroform, followed by back-extraction into hydrochloric acid.[9] Samples were analysed using high resolution ICP-MS.

Quality Control

CSL is United Kingdom Accreditation Service (UKAS) accredited for the methods used for total arsenic analysis and carried out work to criteria considered acceptable by UKAS for inorganic arsenic analysis. Throughout the duration of the study CSL took part in proficiency testing exercises for both total and inorganic arsenic and performed satisfactorily overall.

The quality control criteria used were as follows:

- Results of duplicate analysis were accepted if they had Relative Standard Deviations of less than 25 per cent, or differed by no more than 2 times the limit of detection, whichever was greater.
- Procedural blanks and spiked procedural blanks were analysed with the test samples to estimate recovery. Data were accepted if spike recovery was between 80 and 120 per cent.
- Four Certified Reference Materials (CRMs) were used throughout the survey: NIST 1547 peach leaves, NIST 1549 Non-fat milk powder, DOLT-2 Dogfish liver and NRCC DORM-2 Dogfish muscle. Results for each batch had to be within the certified range, or 25 per cent of the quoted value, whichever was greater.

Results

There was generally good agreement between the results for total arsenic achieved using the two methods (direct nebulisation ICP-MS and hydride generation ICP-MS) for all food groups. A summary of the results for total arsenic as measured by direct nebulisation ICP-MS is given in **Table 1**. The limits of detection (LOD), range and mean arsenic concentrations are given for each food group.

Arsenic was detected in all samples of carcass meat, offal, fish and the other vegetables food groups, and in most samples (approximately three quarters) of the other food groups. The highest concentrations were found in the fish group, with concentrations ranging from 1.1 milligrams/kilogram to 8.4 milligrams/kilogram. The mean arsenic concentration for the fish group of 3.2 milligrams/kilogram is slightly lower than that reported for the 1997 TDS of 4.4 milligrams/kilogram. The second highest concentrations were seen in the poultry group, although the concentrations reported were considerably lower than those seen for

the fish group. The mean arsenic concentration for this group (73 micrograms/kilogram) was higher than that reported in 1997 (4 micrograms/kilogram). This increase is partly due to the analytical method used (direct nebulisation ICP-MS). This method gave similar results to hydride generation ICP-MS for all food groups apart from fish and poultry. This is because fish and poultry contain some organic arsenic species which do not appear to be detected by hydride generation ICP-MS, which was used by previous TDS surveys for all food groups except fish, so would have under-reported the concentration of arsenic in the poultry group.

Mean arsenic concentrations in all other food groups were below 10 micrograms/kilogram, with the exception of the miscellaneous cereals group which contained an average of 13 micrograms/kilogram. These results are similar to previous TDS surveys.

Only samples of the Poultry, Fish and Miscellaneous Cereals food groups were analysed for inorganic arsenic, because the mean levels of total arsenic for all other groups were less than the LOD for inorganic arsenic (10 micrograms per kilogram). A summary of inorganic arsenic concentrations in food groups from the 1999 TDS is also given in **Table 1,** with those values corresponding to inorganic arsenic measurements in brackets.

Inorganic arsenic was detected in the majority of fish samples (20 out of 24), with an upper bound mean concentration of 15.9 micrograms/kilogram. This is less than 1 per cent of the mean concentration of total arsenic recorded. Inorganic arsenic was detectable in less than half (10 out of 24) of the poultry samples, and one eighth (3 out of 24) of the miscellaneous cereal samples. Mean inorganic arsenic concentrations in these groups were 12.5 micrograms/kilogram and 11.6 micrograms/kilogram respectively. Although a greater percentage of the arsenic in these foods is in the form of inorganic arsenic in comparison to fish, total arsenic concentrations in these foods are much lower.

Dietary exposure estimates

Two different types of exposures have been estimated for this survey:

Population exposure estimates are estimated using information on household consumption data from the UK National Food Survey (NFS). The NFS is carried out annually, and the foods that make up the TDS and their relative proportions are based on

three previous years of the NFS. This type of exposure estimate is used to identify trends in exposure, but does not give information on the exposures of individual consumers; and ***Consumer exposure estimates*** take into account exposures by individuals rather than the population as a whole, and also consider those who eat above average amounts of food (97.5th percentile consumers). These estimates therefore provide more accurate assessments of dietary exposures of individual consumers, and are used for comparison with safety guidelines to assess any health implications for consumers. Consumer exposure estimates are based on consumption data from the National Diet and Nutrition surveys (NDNS)[10-14]. Given the detailed nature of the NDNS, it is not possible to up-date them for each age/population group as frequently as the NFS and so they are less suitable for following trends in exposure than population estimates.

Population dietary exposures were estimated by multiplying the amounts of foods consumed based on consumption data from the National Food Survey[6-8] by the corresponding mean concentrations of metals and other elements detected in each TDS food group. Population exposures, expressed on a milligram per day basis are given in **Table 2**.

Dietary exposures for the general UK population to total arsenic estimated from the results of the 1999 TDS are slightly lower than those from the 1994[15] and 1997 TDS (0.05 milligrams per day compared to 0.063 and 0.065 milligrams per day for the 1994 and 1997 TDS respectively). Fish is the main contributor to arsenic in the diet, accounting for 87 per cent of population dietary exposure. This is slightly lower than in 1997, when the fish group contributed 94 per cent of dietary exposure to arsenic. This decrease is because of the lower arsenic concentration reported for the fish group compared to the previous survey. Other food groups contributed far less to dietary intake of total arsenic, with the poultry and the miscellaneous cereals groups each contributing 3 per cent. All other food groups contributed 1 per cent or less to overall exposure. The mean population exposure to inorganic arsenic was between 0.0009-0.005 milligrams per day (exposure was estimated using lower bound mean and upper bound mean, as described previously).

Mean and high-level consumer exposures to total arsenic and inorganic arsenic were estimated for toddlers ($1^1/_2$ - $4^1/_2$ year olds), young people (4-18 year olds), adults, the elderly and for vegetarians (including some fish eaters). A summary of these exposures is given in **Table 3**. Exposures are expressed as a range from lower bound mean (for those

results less than the limit of detection, it has been assumed that concentrations are zero) to upper bound mean (for those results less than the limit of detection, it has been assumed that concentrations are equal to the limit of detection for total arsenic, and for inorganic arsenic assumed to be equal to the concentration of total arsenic). They are expressed on a microgram/kilogram bodyweight basis, which allows for easy comparison to the safety guideline.

Mean adult consumer exposure to total arsenic has decreased from the 1997 TDS. Exposure for the 1999 TDS is 1.3 micrograms/kilogram bodyweight/day compared to the previous estimate which is equivalent to 1.7 micrograms/kilogram bodyweight/day for a 70.1 kilogram adult (reported as 0.12 milligrams/day). High level adult consumer exposure has also decreased (4.4 micrograms/kilogram bodyweight/day compared to the previous estimate which is equivalent to 6 micrograms/kilogram bodyweight/day for a 70.1 kilogram adult (reported as 0.42 milligrams/day)). Adult exposures to inorganic arsenic were much lower. Mean adult exposure ranged from 0.02 - 0.08 micrograms/kilogram bodyweight/day, and high level exposure from 0.05 - 0.1 micrograms/kilogram bodyweight/day.

Exposure estimates for children were higher than those for adults as expected given their higher food consumption on a bodyweight basis. Exposures to total arsenic for vegetarian consumers were the lowest estimated and are probably due to lower/non-consumption of the food groups containing the highest concentration of arsenic, fish and poultry.

Review of the results by the COT- what results mean for consumers.

The COT was asked to consider the results of this survey. It considered that there are no relevant safety guidelines by which to assess the safety of either inorganic or organic arsenic in the diet. Inorganic arsenic is genotoxic and a known human carcinogen and therefore exposure to inorganic arsenic should be as low as reasonably practicable. In this respect, the COT considered that the decreases in adult consumer and population dietary exposures to total arsenic offer reassurance that levels of arsenic in food are not increasing. The COT noted that fish is a major contributor to dietary exposure to arsenic with the predominant form of arsenic in fish being organic. Members also noted that the general assumption that organic arsenic is less toxic than inorganic arsenic is based on an

extremely limited database. However they considered that there is no evidence that exposure to organic arsenic through high levels of fish consumption would result in harmful effects, and therefore concluded that the dietary exposure to organic arsenic identified in the survey was unlikely to constitute a hazard to health. The full COT statement can be accessed at:
www.food.gov.uk/science/ouradvisors/toxicity/statements/cotstatements2003/arsenicstatement

References

1. Ministry of Agriculture, Fisheries and Food (1999). 1997 Total Diet Study: Metals and Other Elements. Food Surveillance Information Sheet No. 191. The Stationery Office, London.

2. World Health Organization (1989). Evaluation of Certain Food Additives and Contaminants. Thirty-third Report of the Joint FAO/WHO Expert Committee on Food Additives. WHO Technical Report Series No. 776. World Health Organization, Geneva.

3. Edmonds, J.S. and Francesconi, K. A. (1993). Arsenic in seafoods: human health aspects and regulations. Marine Pollution, 26, 665-674.

4. Kohlmeyer U, Kuballa J. and Jantzen E. (2002). Simultaneous Separation of 17 inorganic and organic arsenic compounds in marine biota by means of high performance liquid chromatography/inductively coupled plasma mass spectrometry. Rapid Communications in Mass Spectrometry, 16, 965-974.

5. Peattie, M.E., Buss, D.H., Lindsay, D.G. and Smart, G.Q. (1983). Reorganisation of the British Total Diet Study for monitoring food constituents from 1981. Food and Chemical Toxicology, 21, 503-507

6. Ministry of Agriculture, Fisheries and Food (1997). National Food Survey 1996. Annual reports of the National Food Survey Committee. The Stationery Office, London.

7. Ministry of Agriculture, Fisheries and Food (1998). National Food Survey 1997. Annual reports of the National Food Survey Committee. The Stationery Office, London.

8. Ministry of Agriculture, Fisheries and Food (1999). National Food Survey 1998. Annual reports of the National Food Survey Committee. The Stationery Office, London.

9. Munoz, O., Velez, D. and Montoro, R. (1999). Optimisation of the solubilisation, extraction and determination of inorganic arsenic [As III + AsV] in seafood products by acid digestion, solvent extraction and hydride generation atomic absorption spectrometry, The Analyst, 124, 601-607.

10. Gregory, J., Foster, K, Tyler, H and Wiseman, M (1990). Dietary and Nutritional Survey of British Adults. The Stationary Office, London

11. Ministry of Agriculture, Fisheries and Food (March 1999). Vegetarians Dietary Survey: Technical Report on Weighed Intake Diary Data. Research & Development and Surveillance Report:

12. Finch, S., Doyle, W., Lowe, C., Bates, CJ., Prentice, A., Smithers, G. & Clarke PC. (1998). National Diet and Nutrition Survey: People aged 65 years and over. Volume 1 report of the diet and nutrition survey. The Stationery Office.

13. Gregory, J., Lowe, S., Bates, CJ., Prentice, A., Jackson, LV., Smithers, G., Wenlock, R. & Farron M. (2000). National Diet and Nutrition Survey: Young people aged 4 to 18 years, Volume 1: Report of the diet and nutrition survey, The Stationery Office

14. Gregory, J., Collins, DL., Davies, PSW., Hughes, JM. & Clarke, PC. (1995). National Diet and Nutrition Survey; Children Aged 1½ - 4½ Years. Volume 1: Report of the diet and nutrition survey. HMSO.

15. Ministry of Agriculture, Fisheries and Food (1997). 1994 Total Diet Study: Metals and Other Elements. Food Surveillance Information Sheet No. 131.

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A copy of the final report of this survey has been placed in the FSA Library - address detailed above. If you wish to consult a copy, please contact the library for an appointment giving at least 24 hours notice or, alternatively, copies can be obtained from the Library: a charge will be made to cover photocopying and postage.

Table 1. Summary of arsenic concentrations

Food Group	Range of Total Arsenic Concentrations[1] (Range of Inorganic Arsenic Concentrations) (µg/kg)	Total Arsenic Lower Bound Mean (Inorganic Arsenic Lower Bound Mean) (µg/kg)	Total Arsenic Upper Bound Mean (Inorganic Arsenic Upper Bound Mean) (µg/kg)	Total Arsenic LOD (Inorganic Arsenic LOD) (µg/kg)
Bread	<2.1 – 7.5	4.2	4.6	2.1
Miscellaneous cereals	3.2 – 26 (<10 – 19)	13 (5.75)	13 (11.6)	2.1 (10)
Carcass meat	1.6 - 15	5.1	5.1	1.4
Offal	2.2 – 30	10	10	1.4
Meat products	<2.1 – 8.2	3.8	4.1	2.1
Poultry	<2.1 – 169 (<10 – 56)	73 (3.75)	73.1 (12.5)	2.1 (10)
Fish	1106 – 8423 (<10 – 28)	3214 (14.25)	3214 (15.9)	1.4 (10)
Oils and fats	<4.3 – 9.1	2.5	5.1	4.3
Eggs	<1.4 – 7.6	3.1	3.5	1.4
Sugars and preserves	<4.3 – 16	5.8	7.3	4.3
Green vegetables	<0.5 – 6.3	2.0	2.0	0.5
Potatoes	<1.4 – 18	2.1	2.7	1.4
Other vegetables	2.7 – 7.4	4.9	4.9	0.9
Canned vegetables	<0.9 – 2.5	1.6	1.6	0.9
Fresh fruit	<0.9 – 3.7	1.4	1.6	0.9
Fruit products	<0.9 – 5.5	1.9	2.0	0.9
Beverages	<0.5 – 3.0	0.44	0.74	0.5
Milk	<0.5 – 0.7	0.14	0.51	0.5
Dairy produce	<2.1 – 3.4	0.5	2.2	2.1
Nuts	<4.3 - 11	1.7	4.8	4.3

[1] Maximum and minimum values shown are the highest and the lowest reported figures respectively for each food group. Where the minimum value is below the LOD this is indicated (ie <2.1).
Values in brackets represent inorganic arsenic concentrations. The determination of inorganic arsenic was only carried out on the above three food groups because the mean levels of total arsenic in all other food groups were lower than the LOD for inorganic arsenic of 10 µg/kg.

Table 2. Population dietary exposures (milligram/day) to total and inorganic arsenic estimated from the 1999 UK Total Diet Study

	Population dietary exposure to total arsenic from 1999 TDS (mg/day)	Population dietary exposure to inorganic arsenic from 1999 TDS (mg/day)
Total	0.05	0.0009-0.005

Exposures expressed as a range from lower to upper bound. Where the difference between the lower bound and upper bound mean concentrations is very small, rounding of the data leads to a single value.

Table 3. Consumer dietary exposures (microgram//kilogram bodyweight/day) to total and inorganic arsenic estimated from the 1999 Total Diet Study

Population Group	Estimated total dietary exposure to arsenic (µg/kilogram bodyweight/day) *			
	Average		High level	
	Total Arsenic	Inorganic Arsenic	Total Arsenic	Inorganic Arsenic
Adults	1.3	0.02 – 0.08	4.4	0.05 – 0.1
Toddlers (1.5-4.5 years)	2.4 - 2.5	0.05 – 0.2	11.3	0.1 – 0.3
Young people (4-18 years)	1.6	0.03 – 0.1	6.7	0.08 – 0.2
Elderly (free living)	1.6	0.02 – 0.07	5.3	0.04 – 0.1
Elderly (Institutional)	1.4 - 1.5	0.02 – 0.09	4.6	0.05 -0.1
Vegetarians	1.2 – 1.3	0.02 – 0.07	7	0.05 – 0.1

*****Exposures to total and inorganic arsenic have been estimated from a range (lower – upper bound) of mean concentrations. Where the difference between the lower bound and upper bound mean concentrations is very small, rounding of the data leads to a single value.

1. Consumption data taken from the relevant National Diet and Nutritional Surveys [10, 11,12,13,14]

2. The exposure to arsenic by the average consumer and high level (97.5 per centile) consumer for all foods combined is not equal to the sum of the exposure from the individual food. It refers to the dietary exposure by a consumer consuming one or any combination of the foods containing arsenic. These values are derived from a distribution of the individual consumer's consumption patterns with regards to the individual foods.