Monitoring Systems for the Assessment of Dietary Intakes of Contaminants

G.K. GHEORGHIEV

1 INTRODUCTION

Surveillance of chemicals in food is a priority of national authorities and international organisations. In this paper, the following definition is accepted: "Monitoring is a system of repeated observation, measurement, and evaluation for a definite purpose carried out on samples representative of individual foods or the diet in a country, or a given area within the country" (FAO/WHO, 1979).

The data generated from a monitoring system can be used for a number of purposes (FAO/WHO, 1979): (1) to prevent contaminated food from reaching the customer; (2) to estimate the intake of contaminants via food; (3) to indicate the need for, or the effect of, measures to reduce food contamination or keep it below specified statutory limits (control of pesticide use, animal drugs, hygiene practice in production, processing, environmental pollution); (4) to import and export control; (5) to localise sources of food contamination; (6) to establish or check maximum residue limits for chemicals in food; and (7) to correlate levels in the environment or body fluids.

Two objectives of monitoring are acknowledged: regulation and dietary intake assessment. Regulatory monitoring is directed primarily at compliance with established statutory limits and at revealing situations where these limits are expected to be, or suspected of being, exceeded. Accordingly, sampling is biased toward such situations. Sampling plans may involve classification of products according to the potential to accumulate residues (Van Middelem, 1980) as in the "pesticide surveillance index" (Reed, 1985).

This chapter discusses monitoring of food for the estimation of actual dietary intakes of contaminants to serve as an aid to risk assessment and to the epidemiology of food safety (Bunyan *et al.*, 1984; WHO, 1985; Leviton, 1984). Such monitoring is characterised by randomised representative sampling, and is linked closely to food consumption data.

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2 DESIGN AND SAMPLING

Systems to monitor food contaminants have been developed in a number of countries over the past 20 years, mainly as a result of worldwide concern over pesticide residues in food. Systems usually involve the following basic elements:

(1) *Objectives and scope:* These should be clearly defined and stated at the planning stages of the system;

(2) *Structure:* Available technical support: laboratory facilities and network, trained personnel;

(3) *Sampling:* What contaminant(s), foods, populations, sites, frequency, seasons, availability of recent adequate food consumption data;

(4) *Analytical methodologies:* Standard validated methods, protocol, analytical assurance, presentation format for results; and

(5) *Data management:* Generally via computer support.

The Joint FAO/WHO Food and Feed Contamination Monitoring Programme (FCM) is an excellent exercise summarising the best of present day experience in this field (FAO/WHO, 1978; 1979; 1981; 1982a,b; 1984a), and will be relied upon in the present review.

Three basic approaches for sampling food are used: (1) individual food products; (2) total diet (market basket) studies; and (3) duplicate portion.

The approaches are closely linked to diverse types of food consumption data to calculate intake. The combination of these two sets of information are presented on Table 1 based on the Guidelines for the Study of Dietary Intakes of Chemical Contaminants (WHO, 1985).

Individual food products are the simplest samples to analyse for the presence of contaminants, and have been used extensively to estimate intakes. This approach has several advantages. The relative contribution of each food can be evaluated. The procedure is flexible, since it can incorporate various data for food consumption at national and regional levels and is closely related to the network of food control laboratories organised traditionally in many countries.

Three general types of food and feed sampling are distinguishable (Gough *et al.*, 1978):

(1) Regulatory (i.e., selective) sampling to prevent the introduction of products whose contaminants exceed established tolerances;

(2) Surveillance (i.e., objective) sampling to identify situations suspected of being out of compliance (Duggan *et al.*, 1983; National Pesticide Monitoring Programme, 1971); and

(3) Trend sampling to identify shifts in concentration ranges and in intakes.

Existing laboratory facilities and networks can be used for intake studies if their sampling plans are upgraded to type (c) to provide samples as objective and representative as possible.

The distribution of concentration of contaminants in foods is almost invariably non-Gaussian, a situation that has serious implications in determining sampling plans, sample size, and data treatment (Holden, 1970, 1973; FAO/WHO, 1975, 1978; Horwitz and Howard, 1978; Knutti and Schlatter, 1982). Exponential, logarithmic Chi-square, gamma, negative, and binominal distributions require far more samples to detect statistical significance (Holden, 1970; Knutti and Schlatter, 1982).

The range of foods selected for monitoring may vary widely depending on the objectives or contaminants under study. Thus trends in *per capita* cadmium intake in the USA over a more than 50-year period (1920 to 1975) have been estimated on the basis of nine selected staple foods: dairy, meats, fish, poultry, grain, cereals, potato, vegetables and fruits (Travis and Etnier, 1982). Assuming a level concentration of cadmium, dramatic changes in consumption of these staples have brought a 20 percent decline in total intake between 1945 and 1975 (Travis and Etnier, 1982).

With sufficient understanding about environmental mobility of contaminants, sentinel or indicator foods can be used. A typical case is mercury in fish (Sherlock *et al.*, 1982). Representative, "carrier" foods should be selected carefully. Spiegelhalder *et al.* (1980) studied 2826 samples of foods from the market "under suspicion" for *N*-nitroso compounds and found that beer, meats, and cheese contribute 75 percent of the total intake of 1.1 μg/person/day of nitrosodimethylamine, while these products represented only 34 percent of the total food consumption. In a similar study of the UK market, Gough *et al.* (1978) found lower intakes of NDMA of 1 μg/person/week. Had he included beer among the foods sampled, the results would have agreed very closely with those of the German study (Spiegelhalder *et al.*, 1980).

The basic disadvantage of an individual food commodities approach is that the effect of the cooking on the contaminant and the consequent disposal of waste from culinary preparation and incompletely eaten portions cannot be taken into consideration.

In total diet or market basket studies, foods representative of the diet of an "average" person or age/sex group are purchased, prepared for table consumption either individually or, more often, combined in groups (composites), and analysed for specific contaminants. This approach provides highly accurate results about contaminant levels and about relative contributions of each composite. This approach has been used extensively in many countries to search for pesticides and other industrial chemicals (Beattie *et al.*, 1983; Buss and Lindsay, 1978; FAO/WHO, 1982a,b; NPMP, 1971; Podrebarac, 1984a,b), for minerals and metals (Pennington, 1980; Duggan *et al.*, 1983; Slorach *et al.*, 1983; DeVoos *et al.*, 1984), and for polycyclic aromatic hydrocarbons (Dennis *et al.*, 1983). Originally developed for the study of actual intakes of pesticides, the two oldest monitoring systems in the USA and UK have undergone several revisions aimed at

Table 1. Overview and comparison of food consumption data and sampling approaches for contaminant analysis

Type of food consumption data	Type of sampling for contaminant analysis: Total diet (Market basket)	Individual foods commodities	Duplicate portion
Food diary (including weighed intakes) Individual (and population)	Can be used for individuals and groups. Individual patterns can be revealed.	Useful for home-eaters and people with particular eating habits. The usual sampling for food diary consumption.	Suitable to combine with duplicate diet data. Alleviates burden on individual (household) participation. Storage could affect unstable contaminant and create new ones.
Duplicate diet Individual	Useful for small communities and mass catering where duplicate portions can be obtained.		Very appropriate for individual studies. Gives most accurate results. Expensive. Requires high degree of cooperation and respondent burden. Logistics problems likely to occur.
Dietary recall interview Individual (and population)		Widely used for community mass catering with uniform diet in addition to individual studies. Poor accuracy.	Can be woven into interview surveys.

improved sampling, such as expanded coverage of age groups and addition to the list of contaminants and other trace constituents that have raised concern. The first UK revision (Buss and Lindsay, 1978) improved sampling methods, standardised foods for food preparation into 10 groups, and included in the analysis of toxic metals some trace nutrients and polychlorinated biphenyls (PCB).

The UK reorganisation of 1981 tried to overcome some disadvantages in sampling in the earlier system. The new study included 115 foods (68 in the

Table 1. Continued

Type of food consumption data	Type of sampling for contaminant analysis: Total diet (Market basket)	Individual foods commodities	Duplicate portion
Diet history food frequency Individual	Not useful because quantitative data on consumption not available or reliable.	For large groups and identification of specific foods or suspected carriers of particular contaminations.	Not applicable.
Household food disappearance Populations	Useful for regional studies and particular eaters.	Useful for smaller populations or regions. Can use available information on levels of contaminants and high endemic contamination. Difficult in rural subsistence households.	Not applicable.
National food disappearance Population	For general intake estimates on national scale. Does not include regional and group variations. Can be used for approximate estimates and for identification of potential problems.	The usual combination when average national levels of contaminants are available. For rapid overall assessment and comparisons. Usually the least accurate.	Not applicable.

previous) composited into 20 groups (compared to 10 in the previous). This segregation reflected the latest food habit changes and avoided masking the presence of unusual residues in the minor constituents of the larger groups. The foods are purchased in preselected parts of the country by a market research company thus assuring a geographical representation. Instead of relying on the earlier domestic science colleges located in many areas, the preparation and the analysis of the composites have been centralised in one laboratory (Beattie *et al.*, 1983).

Similar trends towards segregation of food sampling appear in the revision

of the US total diet study of April 1982. The changes were as follows (Pennington, 1983):

(1) The list of foodstuffs was enlarged to 200 foods to obtain information on the contribution of separate foods and to reflect changes in food consumption patterns. The foods are collected in four locations (800 foods per year) and analysed in the central facility of the FDA Kansas City Field Office Laboratory. The four locations are selected to obtain a representative sample of the whole country rather than to seek regional differences, which would require a prohibitive number of samples to obtain statistical significance. The regional distinction has been abandoned in part because increasing complexity of food distribution prevents a clear definition of a regional food (Pennington, 1983). All foods were prepared for immediate consumption and analysed individually for 11 essential minerals and more than 120 chemical contaminants.

(2) The number of age/sex groups has been expanded from 3 (6-month old infants, 2-year old toddlers, and teenage males) to 8 (11-month, 2-years, 14 to 16 year males and females, 25 to 30 adults male and female, and 60 to 65 males and females).

In the usual sampling, the food basket typical of the "greatest eater" (as a rule the 15 to 18 year males) is collected. This is the case of a Dutch study of chemical residues in a market basket purchased in only one city and taking into account seasonal differences in the supply of vegetables (DeVoos *et al.,* 1984).

The duplicate diet approach is a direct sampling technique in which an exact duplicate of food being consumed is obtained and analysed. This method is suitable for the estimation of the intakes of individuals and small groups. It provides the most accurate estimates, because it combines results for each contaminant with the actual food consumed. It is limited to small population groups, however, such as hospital kitchens, children's homes, homes for the elderly, and populations living in areas with varying degrees of environmental pollution (Buchet *et al.,* 1983). In the latter study, 124 duplicate samples were analysed, and no regional differences in trace element intakes were revealed. The authors explained this finding by the absence in the diet of home grown vegetables in the corresponding areas (Brussels, Liege, Charleroi) (Buchet *et al.,* 1983). In another typical study, duplicate diets of 98 persons from two UK coastal towns who reported high fish consumption were analysed (Sherlock *et al.,* 1982). The intake levels of mercury were compared with mercury in hair and blood of the same individuals.

In a way similar to the analysis of total diets made up of groups of composites, meals ready for consumption in the home or mass catering locations (e.g., restaurants or canteens) can be analysed. This situation seems to be the closest to real life conditions, and is appropriate as a double

portion, as used to estimate average national intakes of DDT (Gheorghiev, 1982a,b; Soos, 1969) and other contaminants (FAO/WHO, 1982b). Intakes of total DDT and total BHC calculated on the basis of 60 prepared meals in catering establishments agreed well with intakes calculated from monitoring data of residues of these chemicals in individual foods and average national food disappearance data (Gheorghiev, 1982a,b). In this case, the country has a well developed, centralised food distribution system, with no significant differences in total DDT levels found between foods from various areas of the country, while estimates of the intakes for two countries based on their different food consumption patterns proved quite distinct (Gheorghiev, 1982a,b).

When interpreting the above findings, one should bear in mind that DDT is an ubiquitous contaminant whose presence in food is practically unaffected by cooking. The situation could be different with other types of contaminants, some toxic metals for example. Generally in sampling prepared meals, a large number of analyses is needed to obtain a sample that reflects more than a restricted local situation.

Buchet *et al.* (1983) provide another comparison of the intake estimates by various methods of sampling. They report that 2500 samples of individual food items were analysed for metals, and intakes were calculated with the help of average national food disappearance data. They also indicated that monitoring was done on the metal content of food items consumed by 30 Belgian families. The results of the two evaluations were in good agreement: between 260 to 300 μg/person/day for lead, 45 to 50 μg/person/day for cadmium and 14 μg for mercury. The evaluation of Buchet *et al.* (1983) based on 124 duplicate portions gave 96 μg for lead, 15 for cadmium, and 6.5 for mercury.

Table 2 shows the sampling approaches for intake estimates used by a wide cross-section of participants in the programme (FAO/WHO, 1982b).

The total diet is the most versatile sample for intake estimates. The total diet study is evolving towards a more segregated sampling of foods which are prepared ready for consumption. This is due to efforts to reveal the contribution to total burden of individual foods and care not to overlook unusual contaminants and constituents in foods (Beattie *et al.*, 1983; Buss and Lindsay, 1978; Pennington, 1980, 1983; Slorach *et al.*, 1983). Increased volume and costs lead to centralised preparation and analysis (Beattie *et al.*, 1983; Pennington, 1983). The concept of regional food is becoming less important with developing food distribution systems (Gheorghiev, 1982; Pennington, 1983), but may assume some importance in areas having unique pollution problems and where locally grown food constitutes a large part of the diet. Differences in regional consumption patterns should be considered (Gheorghiev, 1982a,b).

Table 15.2. Design of total intake estimates by countries in the joint FAO/WHO food contaminant monitoring programme

	Austria	Canada	Guatemala	Hungary	Japan	New Zealand	United Kingdom	USA
Weight (kg) per day	1.65	1.03	*	*	1.4	3.31	1.5	2.9
Population group	Adult	Average person	Rural, urban, lower income	Institutional; public catering	Average male	Active young man	Average person	Teenage male
Food items collected	Daily personal consumption	120	Complete meals	Complete meals	90	*	78	117
Alcoholic beverages	Yes	*	*	No	Yes	Yes	No	No
Drinking water	Yes	No	*	*	Yes	*	No	Yes
No. of composites	8	11	*	*	14	8	9	12
Prep before analysis	Home, workplace, restaurant	Kitchen prep., including cooking	According to local custom	Institutional; public catering	Kitchen prep., including cooking	Kitchen prep., including cooking	Kitchen prep., including cooking	Kitchen prep., including cooking

The most suitable sampling for individual diets and small groups is the duplicate portion. It is expensive, and requires a high degree of cooperation from the study population.

Composition of age/sex groups depends on the objectives of the monitoring program. For national estimates, there is a tendency to include more age/sex groups rather than to collect the 2-week diet of the 16 to 19 year old males (Pennington, 1983). Results of the total dietary monitoring for infants, toddlers, and adults did not reveal differences in the total intake of some 50 pesticide chemicals, but there were differences in the intake of arsenic, cadmium, lead, mercury, selenium, and zinc (Podrebarac, 1984a; FAO/WHO, 1982a,b).

Frequency of sampling is usually timed to cover seasonal variations in the supply of certain foods or seasonal variations in contamination patterns or degree. In markets with a constant supply of foods, successive sampling (e.g., 7 to 14 days) is used to reflect typical variety of the average diet (WHO, 1985; Slorach *et al.,* 1983).

Rigorous statistical sampling plans are rarely practicable in intake monitoring to the extent to which they are applied to surveillance (Duggan *et al.,* 1983; FAO/WHO 1982a; National Pesticide Monitoring Programme, 1971) and regulatory monitoring (FAO/WHO, 1984b,c). Many factors contribute to the variability of results combined with the deviation from normal distribution (Holden, 1970, 1973; Knutti and Schlatter, 1982; FAO/WHO, 1984b) and may require a very large sample size. This magnitude usually goes beyond available resources, and the programs are reduced to manageable proportions in each particular case (Beattie *et al.,* 1983; Buss and Lindsay, 1978; FAO/WHO, 1984a,b; Pennington, 1983).

3 ANALYTICAL METHODOLOGIES

The methods of analysis and the approach with which they are used to obtain reliable and comparable data are an integral part of any monitoring system; and the design of the measurement process should be performed jointly by the analyst, the statistician, and the interpreters of the data.

The accuracy of the final dietary intake estimates depends more on (a) reliable food consumption data and (b) sampling error, than on (c) the analytical error of the method (Horwitz and Howard, 1978; FAO/WHO, 1979, 1985). Analytical methodologies introduce substantial variability, which increases at trace concentrations typical in food monitoring. It is worthwhile to review at some length recent efforts to streamline and control analytical performance.

Standardised methods are recommended, but emphasis is on validation rather than blind adherence to standard procedures. The validation process is "a value judgment in which the parameters of the method are compared

with the requirements of the analytical data" (Taylor, 1983). The observation that comparable variability in precision has been obtained with different methods (Holden, 1970, 1973; FAO/WHO, 1982; Crosby, 1984; McKinney, 1984) has prompted the formulation of criteria for acceptable analytical data for the environment in general (Cairns and Rogers, 1983) and food specifically (Boyer *et al.*, 1985). A paper of the American Chemical Society has particularly useful definitions and guidelines.

The validation process involves replicate measurements on spiked surrogates, standard reference materials, comparison with independent methods and methods of known accuracy, and the performance of collaborative studies (Taylor, 1983). The collaborative study is the crucial test of the validation process, and has been used extensively in monitoring (Holden, 1970, 1973; Smart, 1984) and in the FCM Programme (FAO/WHO, 1979, 1981). The activity of the Association of Official Analytical Chemists has produced many fundamental documents in this field (Youden and Steiner, 1975).

A validated method is a requirement for a monitoring programme; however, it is not sufficient for the production of valid data. It is well known that several laboratories using the same method on the same sample show great degrees of variability. A systematic procedure to assure quality of the measurements allows for the selection of specified limits of acceptable variability that should then be used to judge the validity of the data (Taylor, 1983). Analytical Quality Assurance (AQA) is an inseparable part of the monitoring process. Detailed AQA has been run throughout the FCM (FAO/WHO, 1979, 1981), the U.S. Pesticide Monitoring Programme (FAO/WHO, 1974; Sherma, 1979; Duggan *et al.*, 1983; Podrebarac, 1984a,b), and other programmes (Buchet *et al.*, 1983; Dennis *et al.*, 1983; Slorach *et al.*, 1983; DeVoos *et al.*, 1984). The AOAC has published a book on the subject (Garfield, 1984).

Basic principles and criteria for the selection of priority contaminants for monitoring are considered in the FCM documents (FAO/WHO, 1978, 1979; WHO, 1985) and many other materials at the international and national levels (Reed, 1985).

Food is a highly complex matrix with a potential for a variety of interactions among contaminants and food constituents and among contaminants themselves. The formation of *N*-nitroso compounds is a typical example. Concern has been expressed about such common trace constituents of food as nitrite and sulfiting agents. More precise estimates of exposure have been recommended especially for bound nitrite in meat products (NRC, 1981). Exposure to sulfiting agents in the diet should be based on direct analysis of foods, particularly irreversibly bound sulfite that may possibly induce adverse effects when ingested (FASEB, 1984). Thus if dietary exposure assessment is to be relevant to the epidemiology of multifactorial causation of disease (Leviton, 1984), multi-chemical method-

ologies should be used and appropriate information on the chemical composition of food will be needed (Slorach *et al.*, 1983).

Multi-residue methodology for pesticides and other industrial organics is well advanced (FDA, 1982a,b; FAO/WHO, 1984c; Klisenko, 1984). The AOAC method can determine more than 300 chemicals (McMahon and Burke, 1978; FDA, 1982a,b). Validated methods are also available for multi-residue (Klisenko, 1984) and individual pesticides (FDA, 1982a; FAO/WHO, 1984c; Klisenko, 1984). The method of choice for trace elements and heavy metals is atomic absorption spectroscopy (Tanner, 1982; Tsalev and Zaprianov, 1983), and the recently developed inductively coupled plasma atomic emission spectroscopy offers the best perspective for truly multi-element analysis (Tanner, 1982; Fassel, 1984). Mycotoxins will increasingly be determined by high performance liquid chromatography (IARC, 1982; Tanner, 1982; Crosby, 1984) which is rapid and easy to use (Crosby, 1984). IARC has been publishing monographs about carcinogens, which also include references to methods for foods (IARC, 1978a,b, 1979, 1981, 1982, 1983, 1985).

4 DATA MANAGEMENT

A variety of procedures (Buchet *et al.*, 1983; Dennis *et al.*, 1983; DeVoos *et al.*, 1984), discussions and recommendations (WHO, 1985; FAO/WHO, 1978, 1979, 1984b; Sherlock *et al.*, 1982) concerning the presentation of results at or near the limit of detection and quantitation have been published.

Monitoring systems generate a large volume of data that should be manipulated, stored, and retrieved by a suitable computer. Monitoring data should be stored for later comparison and analysis (FAO/WHO, 1979, 1984c). A central computing unit is usually recommended, and if sources are available this can be connected to a network of remote interactive terminals (FAO/WHO, 1979). While participants should be familiar with the work of the computer system, it should be left in the hands of trained professionals in information handling.

5 CONCLUSIONS

Monitoring food to determine actual exposure to chemicals is a process basic to chemical and food safety programmes (Bunyan *et al.*, 1984; Leviton, 1984; FAO/WHO, 1984). Food safety is viewed as an integral part of primary health care; identification and monitoring of critical points in the food chain is high on the list of recommendations (FAO/WHO, 1984). Monitoring thus can be related to broader programmes. Cost-effective planning and design of sampling and analysis are essential.

Sampling is well defined in three main approaches of increasing accuracy of intake estimates: individual food commodities, total diet study, and the duplicate portion study. Complexity and the cost of performing these studies increase in the same order.

The measuring process of contaminants in foods has developed into a system involving method validation collaborative studies, quality assurance and criteria for acceptance of methods and data. The analysis of chemicals in trace quantities with a specified reliability requires sophisticated instrumentation and highly skilled analysts. As an extreme example, Horwitz (1983, 1984) has estimated that the analysis of dioxin in fish at 50 ng/kg would cost at least $1000 (US) per sample! The cost is certainly lower for more common contaminants, and generally simpler methods of food monitoring are ineffective.

Given the present state of risk assessment, one might ask: what would be the improvement in risk estimates using more precise dietary intake estimates? Actual daily intakes of DDT, dieldrin, nitrosodimethylamine, and aflatoxin B1 for various countries have been compared (Gheorghiev, 1982a,b) with "virtually safe doses" (VSD) obtained by extrapolation by four widely used models: Armitage-Doll, Weibull, one-hit and multi-hit (Food Safety Council, 1982). The VSD's were much lower than existing regulatory limits and acceptable daily intakes for these chemicals. All actual intakes were well within the area of VSD bracketed by the most (one-hit) and least (multi-hit) extrapolation methods.

One might also ask whether the costs are acceptable for a well designed total diet study to check if the dietary intake of pesticide residues in Finland (calculated to be approximately 150 μg/person on the basis of analysis of individual foods (Heminki *et al.*, 1983)) is really 3 times that of the adult US citizen (determined to be 50 μg/person by the most elaborate diet study (Podrebarac, 1984b)). In other cases, exposure estimates are so unreliable that no decision about the safety or otherwise can be made (National Research Council, 1981).

The relevance of monitoring results is closely related to improving our understanding and interpretation of the low and very low dietary intakes of trace chemicals. All monitoring efforts should be very carefully balanced, planned and organised by the experts and the users.

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